(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: 0 298 196 A1

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 88103885.5

(22) Date of filing: 11.03.88

(51) Int. Cl.4: C07D 233/90 , C07D 233/92 , C07D 405/04 , C07D 409/04 , C07D 401/04 , C07D 401/12 , C07D 409/12 , C07D 405/14 , C07F 7/08 , A01N 43/56 , A61K 31/415 , //C07D233/64, C07D233/58

The title of the invention has been amended (Guidelines for Examination in the EPO, A-III, 7.3).

Claims for the following Contracting State: ES.

(30) Priority: 13.03.87 JP 58451/87
03.04.87 JP 82546/87
30.04.87 JP 106577/87

(43) Date of publication of application:
11.01.89 Bulletin 89/02

(84) Designated Contracting States:
AT BE CH DE ES FR GB GR IT LI LU NL SE

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EP 0 298 196 A1

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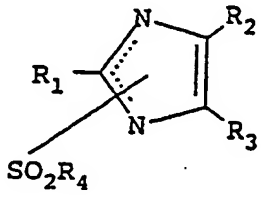
(54) **Imidazole compounds and biocidal compositions comprising the same.**

(57) Imidazole compounds represented by general formula I:



(I)

wherein:

R_1 represents a cyano group or a -CSNHR$_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a -COR$_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; an optionally substituted C_{1-12} alkyl, C_{2-10} alkenyl, C_{1-6} alkoxy, phenyl, furyl, thionyl or pyridyl group; an-SO$_n$R$_7$ group, wherein R_7 represents a C_{1-6} alkyl, C_{2-6} alkenyl , benzyl, an optionally substituted phenyl or pyridyl group; an - NR$_8$R$_9$ group, group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a -CO(NH)$_m$R$_{10}$ gorup, wherein R_{10} represents an optionally substituted C_{1-4} alkyl, C_{1-4} alkoxy or phenyl group and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an -NR$_{11}$R$_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom. The compounds are effective as biocides.

IMIDAZOLE COMPOUNDS AND BIOCIDAL COMPOSITION COMPRISING THE SAME FOR CONTROLLING HARMFUL ORGANISMS

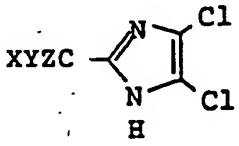
FIELD OF THE INVENTION

The present invention relates to novel imidazole compounds and biocidal compositions comprising the same for controlling harmful organisms.

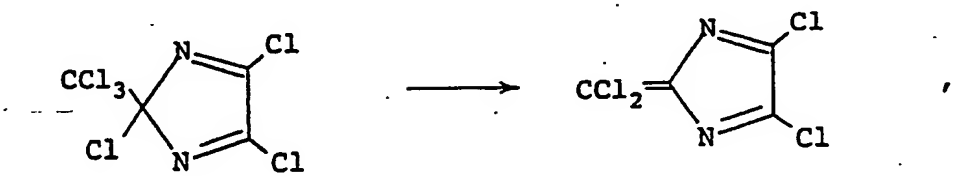
BACKGROUND OF THE INVENTION

Imidazole type compounds proposed so far are exemplified below.

Belgian Patent 852313 (published Sept. 12, 1977) discloses (4,5)-dichloro-imidazole(2)-carboxylic acid derivatives having the formula

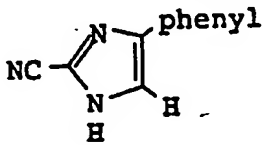
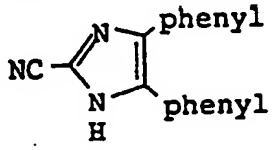


wherein CXYZ represents a C atom with 3 bonds attached to hetero atoms, and Japanese Patent Publication No. 15625/85 (published Apr. 20, 1985) discloses the following reaction scheme,

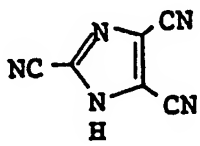
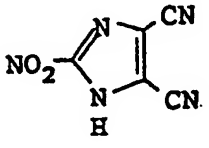


while no compound having other substituents than chlorine atoms at the 4 and 5-positions in the imidazole ring and having a substituted sulfonyl group in the imidazole ring is disclosed in both of the above references.

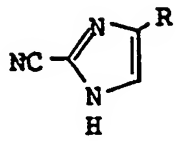
Recl. Trav. Chim. Pays-Bas ,1973, 92(3), 449-59 discloses


,

,

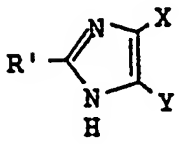
etc.; DT-OS 2317453 (published Oct. 11, 1973) discloses quaternary ammonium salts of


or


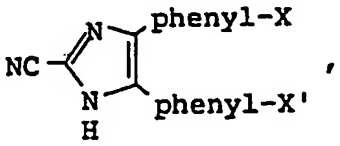
etc.; J. Org. Chem., Vol. 44, No. 16, 1979, 2902-2906 discloses



(R: H, CH_3), etc.; EP 31086 (published July 1, 1981) discloses

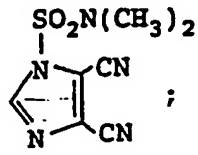


(R': -CQZR, CN); J. Org. Chem., Vol. 51, No. 10, 1986, 1891-1894 discloses 2-cyano imidazole, etc.; and Research Disclosure, June (1986), 323-324 (C.A., 106, 49942e) discloses

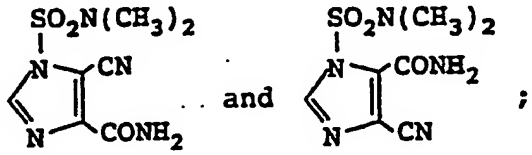


etc.; while no compound having a substituted sulfonyl group in the imidazole ring is disclosed in any of the above-described references.

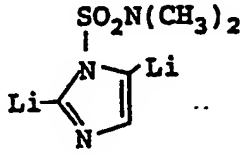
Japanese Patent Application (OPI) No. 4303/80 (published Jan. 12, 1980) (the term "OPI" as used herein means a "published unexamined patent application") discloses 1-(N,N-dimethylsulfamoyl)-4,5-dicyanoimidazole



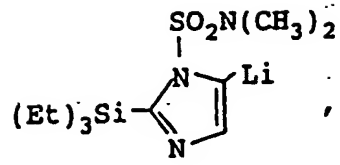
C.A., 95: 7283q [Japanese Patent Application (OPI) No. 157570/80 (published Dec. 8, 1980)] discloses sulfamoylimidazole derivatives of



C.A., 101: 7092u (J.Chem. Soc., Perkin Trans. 1, 1984, (3), 481-6) discloses

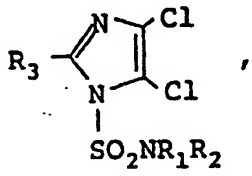


etc.; and C.A., 106: 138324x (Tetrahedron, 1986, 42(8), 2351-8) discloses



etc.; while no compounds having other than a hydrogen atom, a lithium atom, or an -Si(Et)$_3$ group at the 2-position in the imidazole ring as a substituent are disclosed.

Japanese Patent Application (OPI) No. 142164/87 (published June 25, 1987) discloses 4,5-dichloro-imidazole compounds having the formula



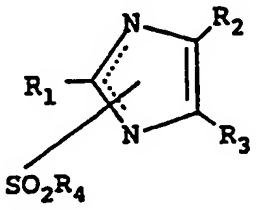
while no compounds having other substituents than chlorine atoms at the 4 and 5-positions in the imidazole ring are disclosed.

References listed below disclose imidazopyridine compounds and/or benzimidazole compounds in which the compounds contain a condensed ring of an imidazole ring with a benzene ring and/or a pyridine ring in their chemical structures.

U.S. Patent 3609157 (issued Sept. 28, 1971)
U.S. Patent 3681369 (issued Aug. 1, 1972)
Belgian patent 830719 (published Dec. 29, 1975)
Belgian Patent 845641 (published Feb. 28, 1977)
U.S. Patent 4536502 (issued Aug. 20, 1985)
U.S. Patent 4579853 (issued Apr. 1, 1986)
French Patent 2559150 (published Aug. 9, 1985)
Japanese Patent Application (OPI) No. 103873/86 (published May 22, 1086)
Japanese Patent Application (OPI) No. 22782/87 (published Jan. 30, 1987)
EP 219192 (published Apr. 22, 1987)
Japanese Patent Application (OPI) No. 195379/87 (published Aug. 28, 1987)
EP 239508 (published Sept. 30, 1987)

SUMMARY OF THE INVENTION

An object of the present invention is to provide imidazole compounds of the following general formula (I) and biocidal compositions comprising the same for controlling harmful organisms:



(I)

wherein:

R_1 represents a cyano group or a -CSNHR$_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a -COR$_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a

phenyl group;

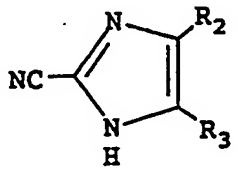
R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted. with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an $-NR_{11}R_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom;

Another object of the present invention is to provide a process for preparing the imidazole compounds of the formula (I) hereinabove.

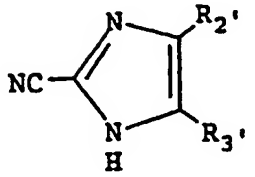
A further object of the present invention is to provide intermediate compounds of the following general formula (II'):



(II')

wherein R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1, provided

that compounds represented by the following general formula (II"):



(II")

wherein R_2' and R_3' are simultaneously a hydrogen atom, a halogen atom, a cyano group, or a phenyl group which is optionally substituted with same or different C_{1-2} alkoxy group or C_{1-2} alkylthio group at the para-position; and wherein R_2' is a hydrogen atom and R_3' is a methyl group or a phenyl group, are excluded.

Among the imidazole compounds represented by the general formula (I), preferred compounds of the present invention are illustrated below.

Compounds of the general formula (I) wherein R_1 is a cyano group;

Compounds of the general formula (I) wherein R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups or nitro groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a phenyl group which is optionally substituted with one or more halogen atoms; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CONHR_{10}$ group, wherein R_{10} represents a phenyl group which is optionally substituted with one or more halogen atoms, provided that R_2 and R_3 are not simultaneously a halogen atom;

Compounds of the general formula (I) wherein R_4 is a C_{1-6} alkyl group or an $-NR_{11}R_{12}$ group, wherein R_{11} and R_{12} each represents a C_{1-4} alkyl group;

Compounds of the general formula (I) wherein R_2 is a hydrogen atom; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, phenyl groups, or halogenated phenyl groups; a C_{2-4} alkenyl group; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, C_{1-4} alkoxy groups, or halogenated C_{1-4} alkoxy groups; a C_{1-6} alkylthio group; or a phenylthio group which is optionally substituted with one or more halogen atoms;

Compounds of the general formula (I) wherein R_3 is a hydrogen atom, a halogen atom, or a cyano group;

Compounds of the general formula (I) wherein R_4 is an $-N(CH_3)_2$ group;

Compounds of the general formula (I) wherein R_2 is a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, phenyl groups, or halogenated phenyl groups; a C_{2-4} alkenyl group; a phenyl group which is optionally substituted with one or more halogen atoms; or a C_{1-6} alkylthio group;

Compounds of the general formula (I) wherein R_3 is a halogen atom; and

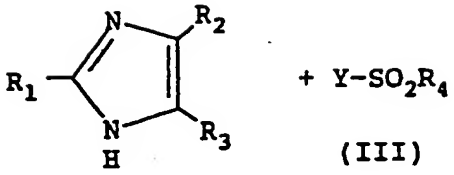
Compounds of the general formula (I) wherein R_1 represents a cyano group; R_2 represents a C_{1-12} alkyl group or a phenyl group; R_3 represents a chlorine atom; and R_4 represents an $-N(CH_3)_2$ group.

DETAILED DESCRIPTION OF THE INVENTION

In the general formula (I) described above, definitions of C_{1-4} alkyl group and alkyl moieties of C_{1-4} alkoxy group and C_{1-4} alkylthio group may include methyl, ethyl, n propyl, isopropyl, n-butyl, sec-butyl, isobutyl and tert-butyl groups. Definition of C_{1-6} alkyl group may include n-pentyl and n-hexyl groups in addition to the exemplified C_{1-4} alkyl groups hereinabove. Definition of C_{1-12} alkyl group may include heptyl, octyl, nonyl, and decyl groups in addition to the exemplified C_{1-6} alkyl groups hereinabove. Definition of C_{3-6} cycloalkyl group may include cyclopropyl, cyclobutyl, cyclopentyl, and cyclohexyl groups. Definition of C_{2-4} alkenyl group may include an allyl group, etc. Definition of C_{2-6} alkenyl group may include a pentenyl group, etc. in addition to the exemplified C_{2-4} alkenyl groups hereinabove. Definition of C_{2-10} alkenyl group may include a geranyl group, etc. in addition to the exemplified C_{2-6} alkenyl groups hereinabove. Definition of halogen atom may include chlorine, bromine, fluorine, and iodine atoms.

The novel imidazole compound represented by the general formula (I) described above can be prepared specifically by the following process:

[A]



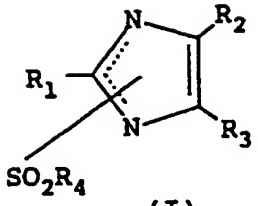
+ $Y-SO_2R_4$

(III)

(II)

10 to 150°C

1 to 48 hours

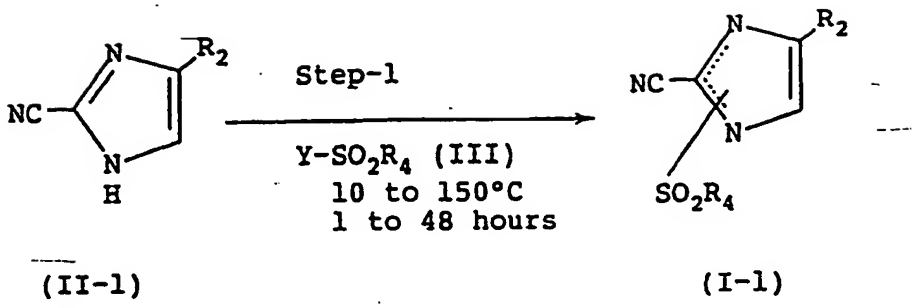


(I)

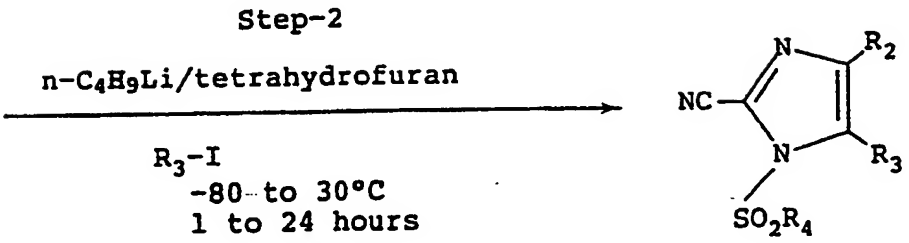
wherein R_1, R_2, R_3, and R_4 have the same meanings as described above; and Y is a halogen atom.

In the general formula (I) described above, compounds wherein R_1 is a cyano group can also be prepared by the following process:

[B]



(II-1) (I-1)

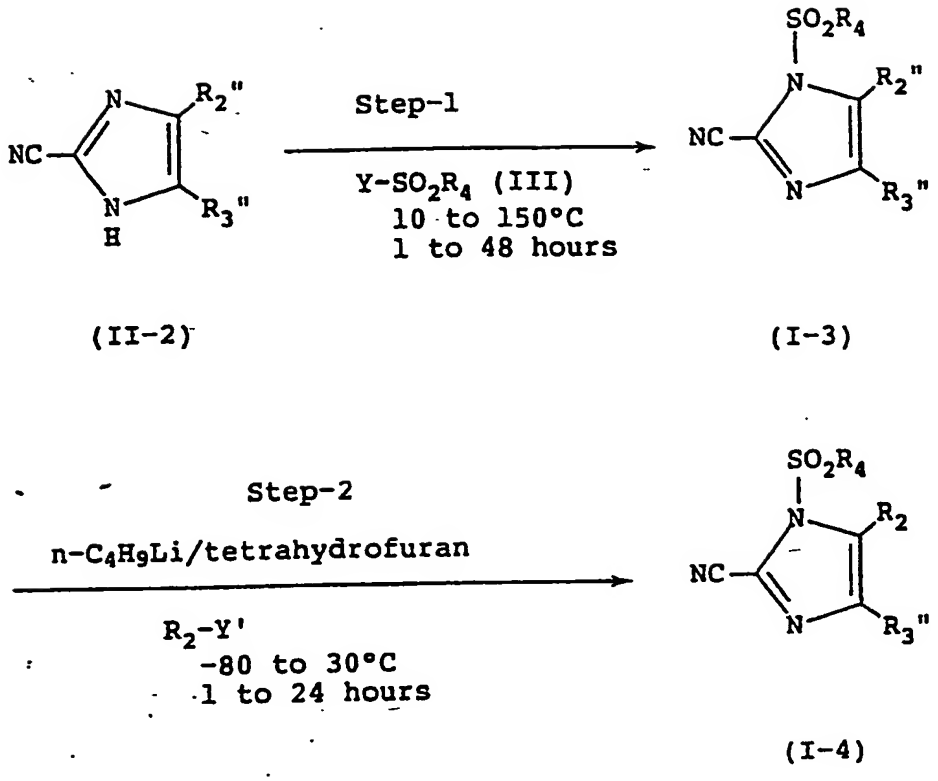


(I-2)

In the general formula (I-2) described above wherein R_3 is an $-SR_7$ group, R_7SSR_7 can also be used instead of R_3-I in Step-2 of the process [B] described above. In the foregoing formulae, R_2, R_3, R_4, R_7, and Y have the same meanings as described above.

In the general formula (I) described above, compounds wherein R_1 is a cyano group, and R_3 is a hydrogen atom, a chlorine atom, or a bromine atom can also be prepared by the following process:

[C]

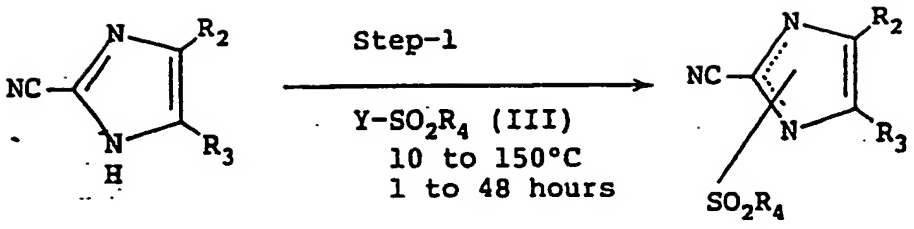


(II-2) (I-3) (I-4)

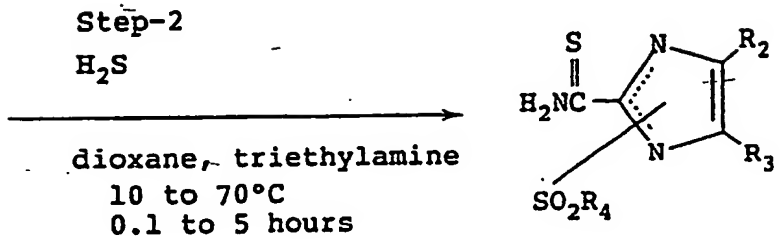
In the general formula (I-4) described above wherein R_2 is an $-SR_7$ group, R_7SSR_7 can also be used instead of R_2-Y' in Step-2 of the process [C] described above; and wherein R_2 is a $-CH(OH)-R_{13}$ group (wherein R_{13} is an alkyl group or an optionally substituted phenyl group), R_{13}-CHO can also be used instead of R_2-Y' in Step-2 of the process [C] described above. In the foregoing formulae, R_2, R_4, and R_7 have the same meanings as described above; R_2'' and R_3'' are simultaneously a hydrogen atom, a chlorine atom or a bromine atom; and Y' is a chlorine atom, a bromine atom, or a iodine atom.

In the general formula (I) described above, compounds wherein R_1 is a $-CSNH_2$ group or a $-CSNHCOR_6$ group can also be prepared by the following process:

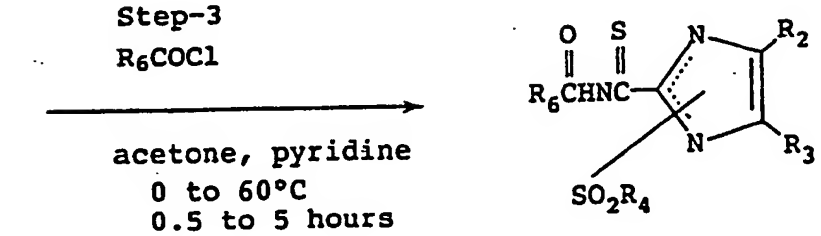
[D]



(II-3) (I-5)



(I-6)



(I-7)

wherein R_2, R_3, R_4, R_5, and Y have the same meanings as described above.

The process [A] and Step-1 of the processes [B] through [D] described above are carried out, if necessary and desired, in the presence of a solvent and an acid acceptor.

Examples of the solvent include aromatic hydrocarbons such as benzene, toluene, xylene, chlorobenzene, etc.; cyclic or acyclic aliphatic hydrocarbons such as chloroform, carbon tetrachloride, methylene chloride, dichloroethane, trichloroethane, n-hexane, cyclohexane, etc.; ethers such as diethyl ether, dioxane, tetrahydrofuran, etc.; ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone, etc.; nitriles such as acetonitrile, propionitrile, etc.; and aprotic polar solvents such as dimethylformamide, N-methylpyrrolidone, dimethyl sulfoxide, sulfolane, etc.

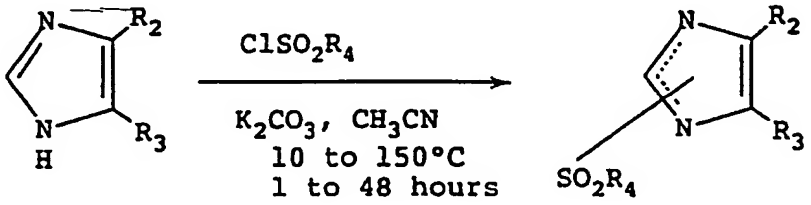
As the acid acceptor, any of inorganic bases and organic bases can be used. Examples of the inorganic base include alkali metal hydroxides such as sodium hydroxide, potassium hydroxide, etc.; alkali metal or alkaline earth metal carbonates such as anhydrous potassium carbonate, anhydrous calcium carbonate, etc.; alkali metal hydrides such as sodium hydride; alkali metals such as metallic sodium; etc. Further, as the organic base, metion may be made of triethylamine, etc.

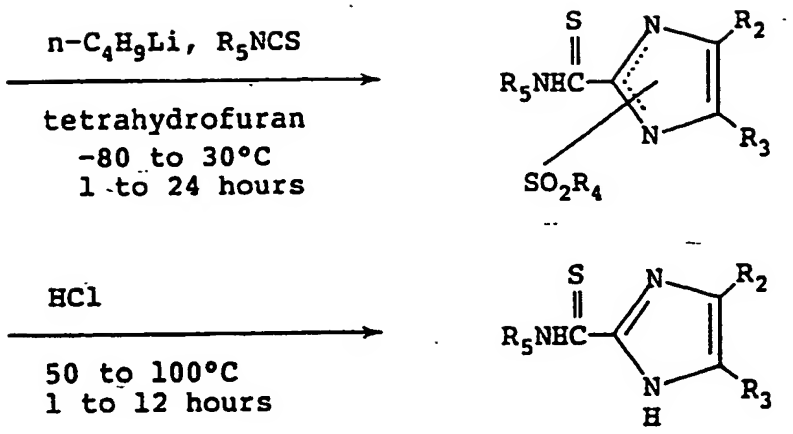
The reaction described above can be carried out in the presence of a suitable catalyst. As the catalyst, mention may be made of, for example, a phase transfer catalyst such as a quaternary ammonium derivative.

As the halongen atom shown by Y in the general formula (III) described above, mention may be made of a chlorine atom, a bromine atom, an iodine atom, and a fluorine atom; of these, preferred is a chlorine atom.

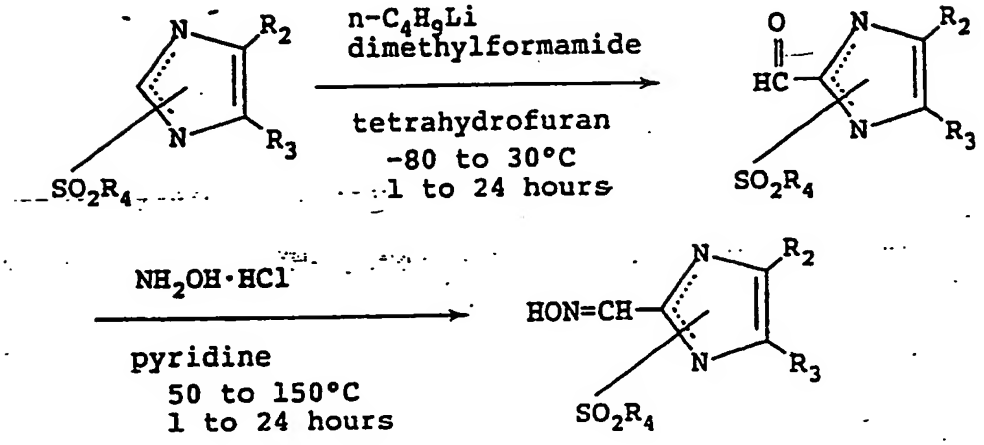
In the reaction scheme described above, the compounds represented by the general formula (III) are known compounds, and the compounds represented by the general formula (II) can be prepared by either one of the following processes.

(1)

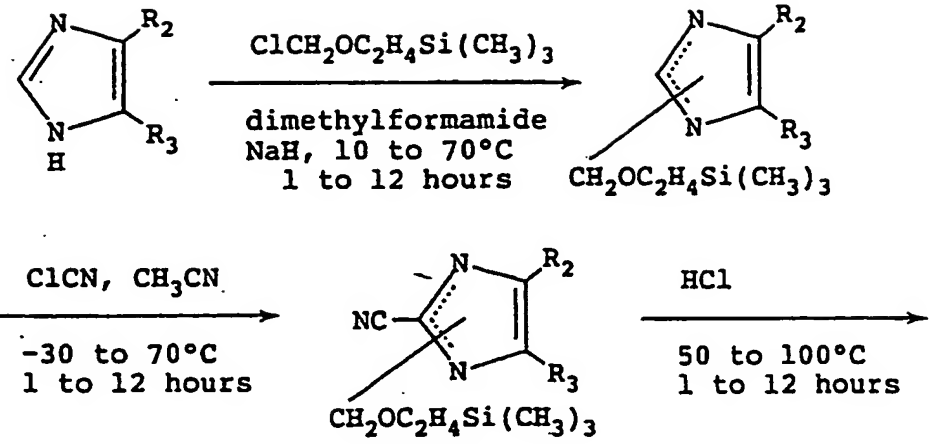



n-C4H9Li, R5NCS
tetrahydrofuran
-80 to 30°C
1 to 24 hours
HCl
50 to 100°C
1 to 12 hours

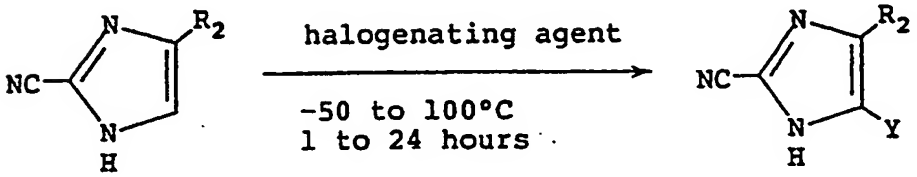
(2)


n-C4H9Li
dimethylformamide
tetrahydrofuran
-80 to 30°C
1 to 24 hours
NH2OH·HCl
pyridine
50 to 150°C
1 to 24 hours
acetic anhydride
pyridine
50 to 150°C
1 to 24 hours
H2S
triethylamine
pyridine
10 to 70°C
0.1 to 5 hours

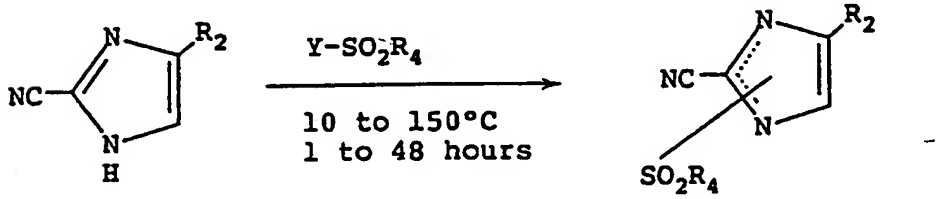
(3)

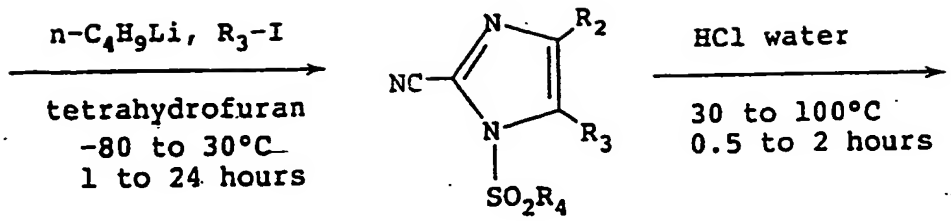

ClCH2OC2H4Si(CH3)3
dimethylformamide
NaH, 10 to 70°C
1 to 12 hours
CH2OC2H4Si(CH3)3
ClCN, CH3CN
-30 to 70°C
1 to 12 hours
HCl
50 to 100°C
1 to 12 hours

(4)

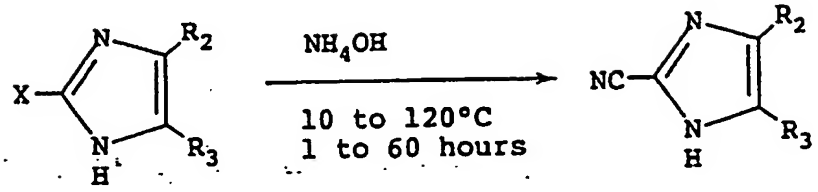

halogenating agent
-50 to 100°C
1 to 24 hours

(5)

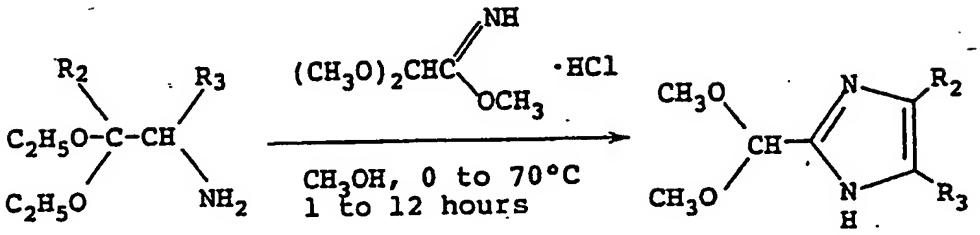

Y-SO2R4
10 to 150°C
1 to 48 hours

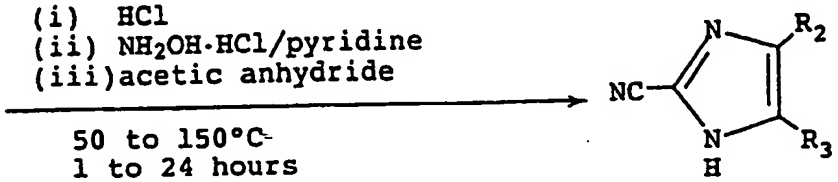

n-C4H9Li, R3-I
tetrahydrofuran
-80 to 30°C
1 to 24 hours
HCl water
30 to 100°C
0.5 to 2 hours

(6)

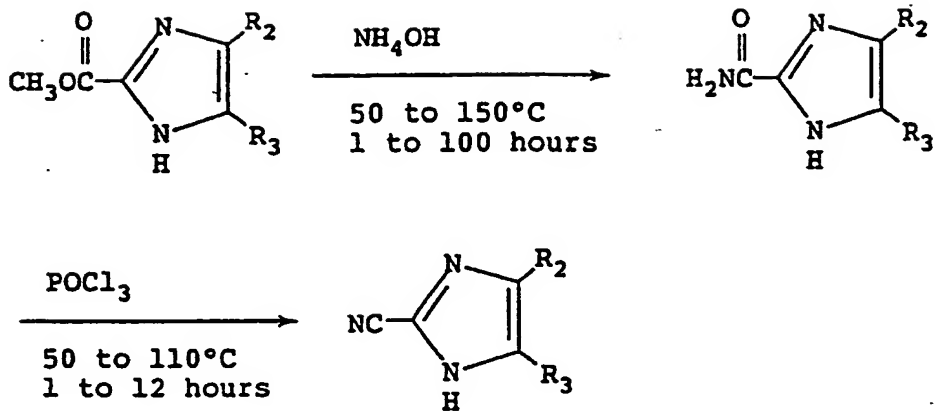

NH4OH
10 to 120°C
1 to 60 hours

(7)

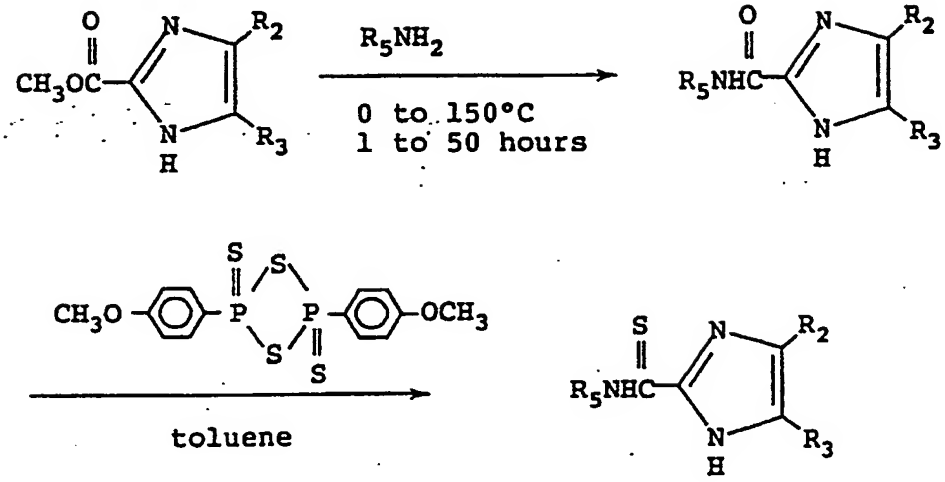

CH3OH, 0 to 70°C
1 to 12 hours


(i) HCl
(ii) NH2OH·HCl/pyridine
(iii)acetic anhydride
50 to 150°C
1 to 24 hours

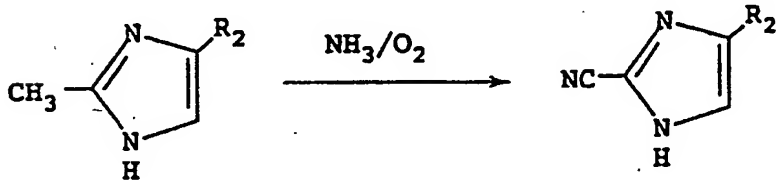
(8)


NH4OH
50 to 150°C
1 to 100 hours
POCl3
50 to 110°C
1 to 12 hours

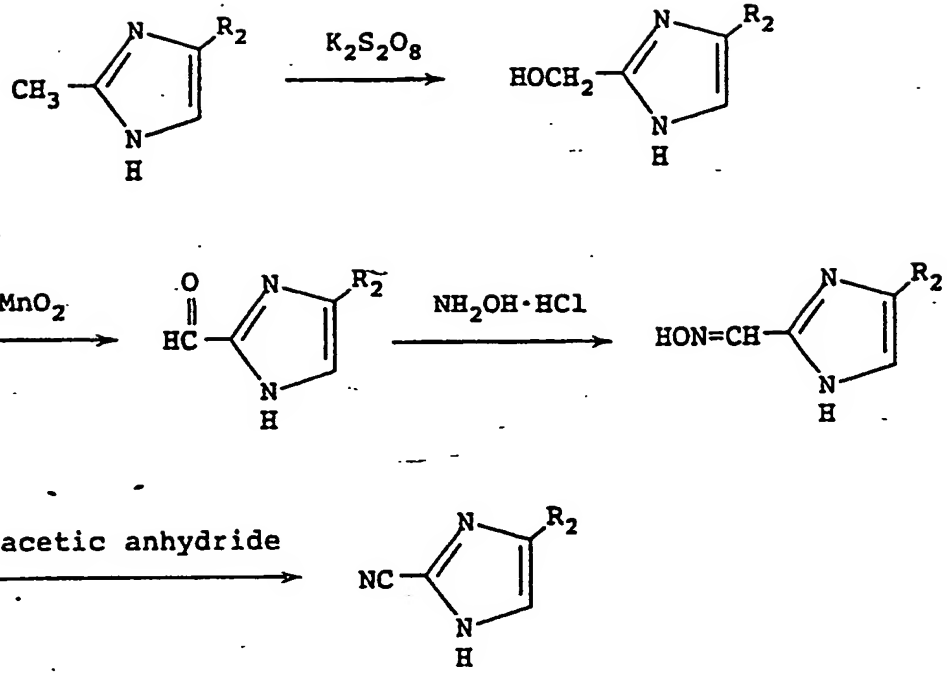
(9)


R5NH2
0 to 150°C
1 to 50 hours
toluene

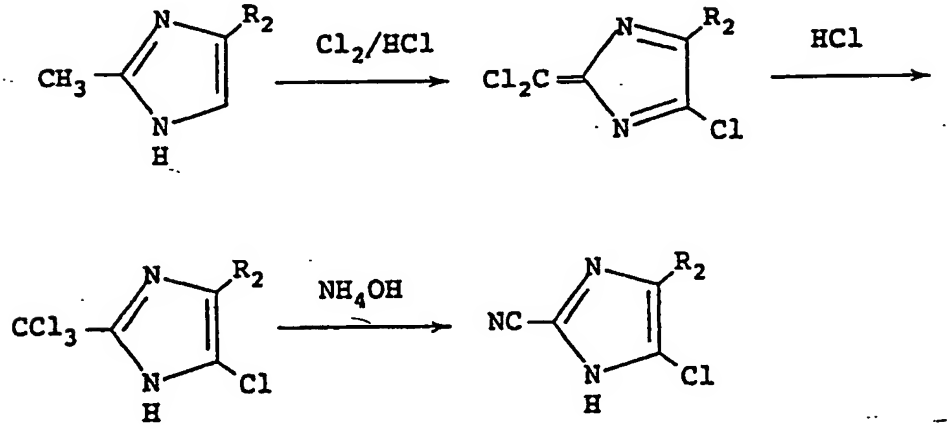
(10)


NH3/O2

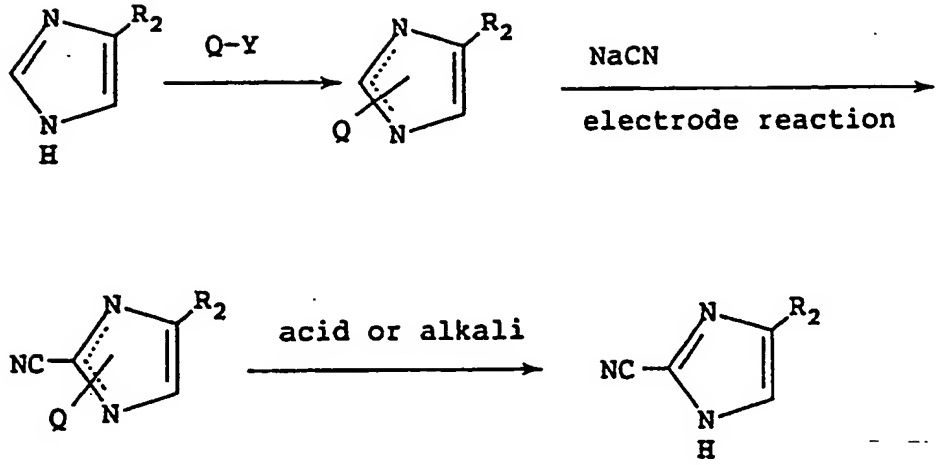
(11)


CH3
N
R2
H
K2S2O8
HOCH2
MnO2
O
HC
NH2OH·HCl
HON=CH
acetic anhydride
NC

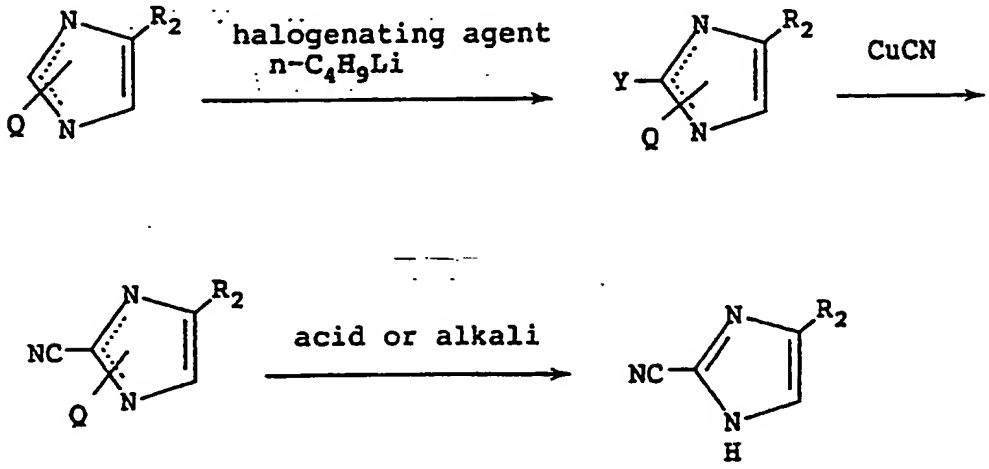
(12)


CH3
Cl2/HCl
Cl2C
HCl
Cl
CCl3
NH4OH
NC

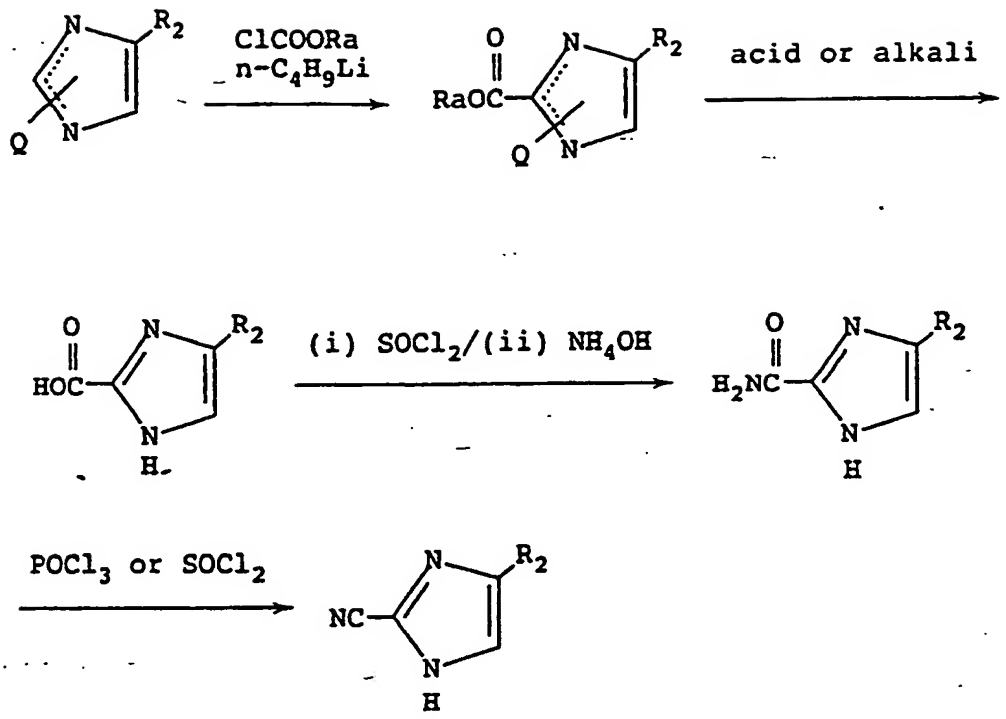
(13)


Q-Y
NaCN
electrode reaction
acid or alkali

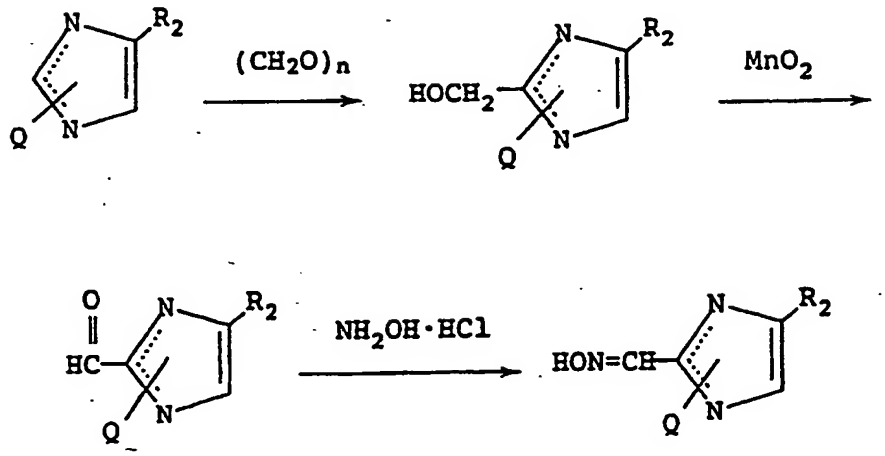
(14)

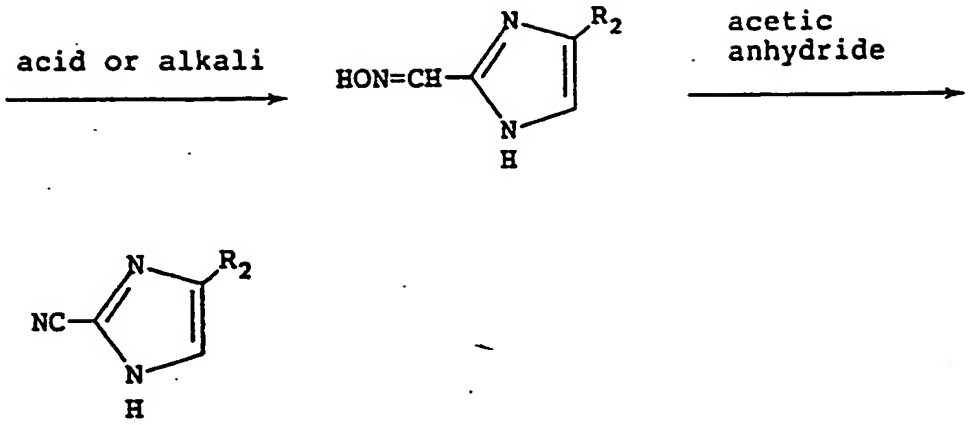

halogenating agent
n-C4H9Li
CuCN
acid or alkali

(15)

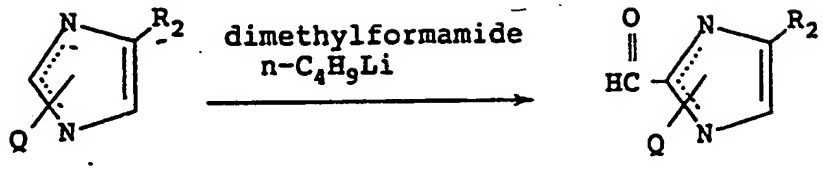

ClCOORa
n-C4H9Li
acid or alkali
(i) SOCl2/(ii) NH4OH
POCl3 or SOCl2

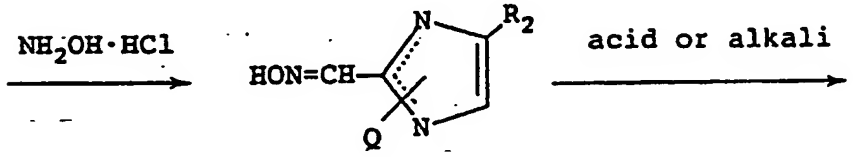
(16)

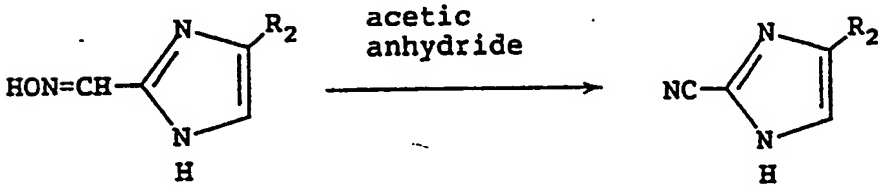

(CH2O)n
MnO2
NH2OH·HCl


acid or alkali
HON=CH
acetic anhydride
NC
H

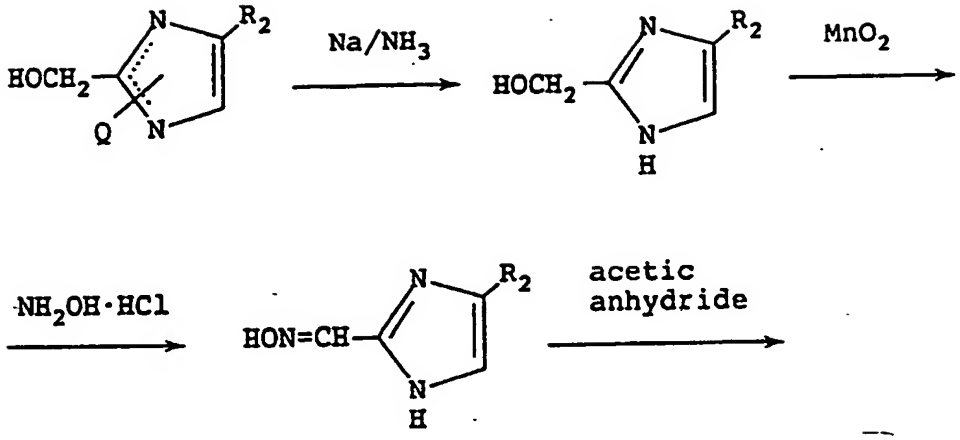
(17)


dimethylformamide
n-C4H9Li
HC
Q

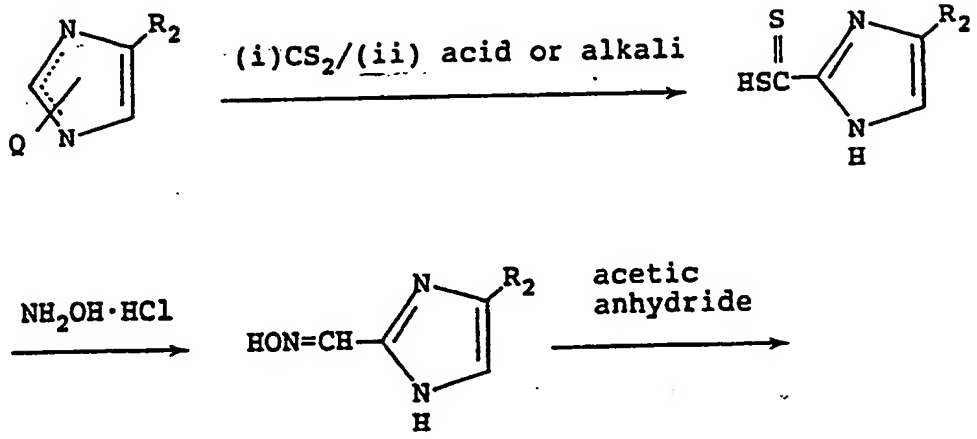

NH2OH·HCl
HON=CH
acid or alkali
Q

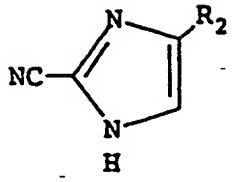

HON=CH
acetic anhydride
NC
H

(18)

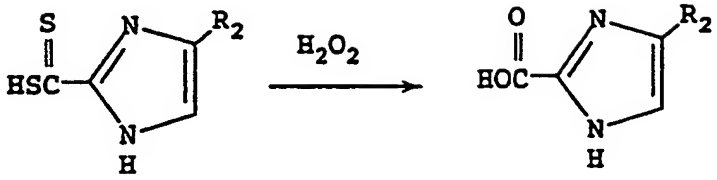

HOCH2
Q
N
R2
Na/NH3
MnO2
NH2OH·HCl
HON=CH
acetic anhydride
NC
H

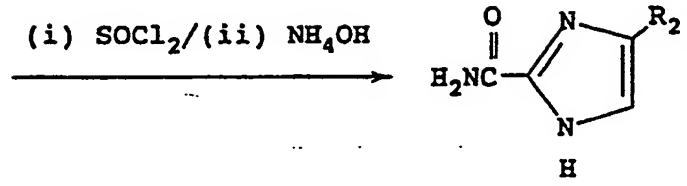
(19)

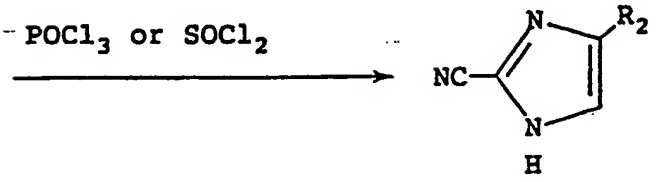

Q
(i)CS2/(ii) acid or alkali
S
HSC
NH2OH·HCl
HON=CH
acetic anhydride



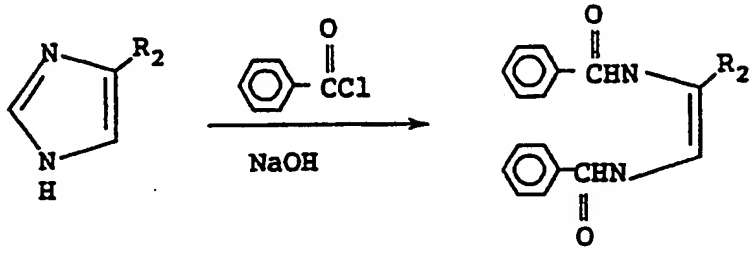
(20)

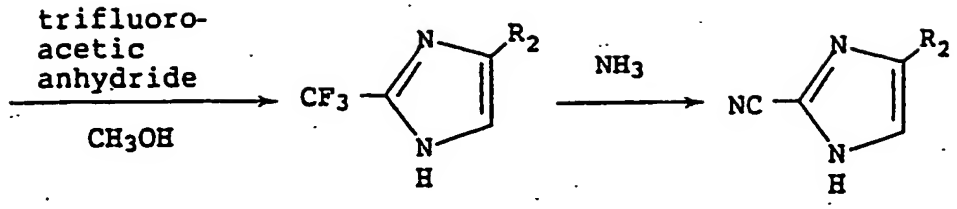




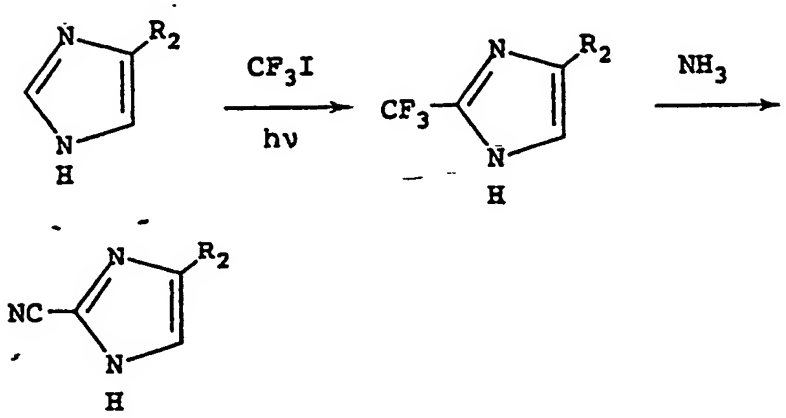


(21)





(22)

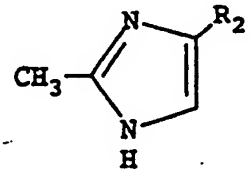


In the foregoing formulae, R_2, R_3, R_4, R_5, and Y have the same meanings as described above; X is a CF_3 group or a CCl_3 group; Ra is an alkyl group; and Q is a protective group.

As the protective groups for Q, an $-SO_2Rb$ group, wherein Rb is a dialkylamino group, an alkyl group, or an optionally alkylated phenyl group; a -CH(Rc)-Rd group, wherein Rc is a hydrogen atom or a methyl group, and Rd is an alkoxy group, a phenyl group which is optionally substituted with an alkyl group or an alkoxy group, or a $-OC_2H_4Si(CH_3)_3$, etc. are exemplified.

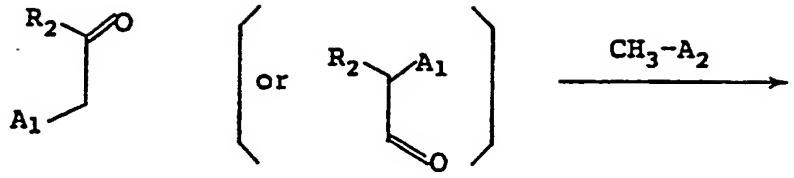
In each of the processes as described above, the reaction conditions such as reaction temperature, reaction time, solvent, acid acceptor, alkali acceptor, etc. can appropriately be chosen from the conventionally known reaction conditions.

Further, the compounds of the formula

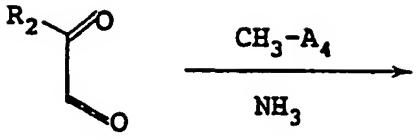


in the reaction schemes of the processes (10), (11), and (12) described above can be prepared by, for example, the following methods:

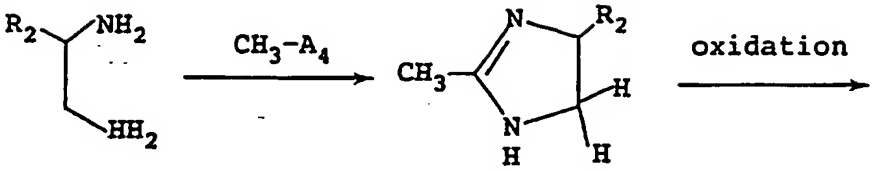
(23)



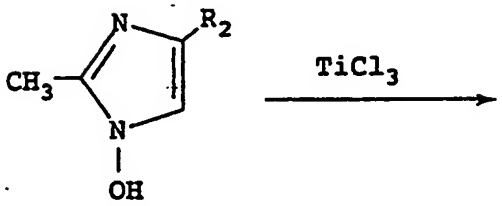
(24)



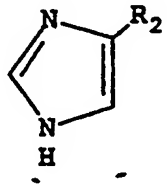
(25)



(26)

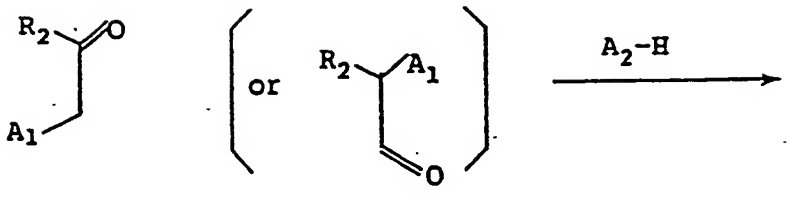


Still further, the compounds of the formula

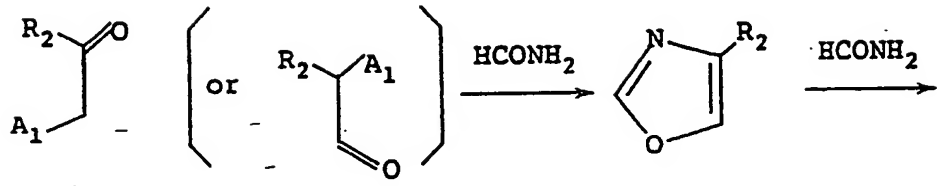


in the reaction schemes of the processes (13), (21), and (22) described above can be prepared by, for example, the following methods:

(27)



(28)



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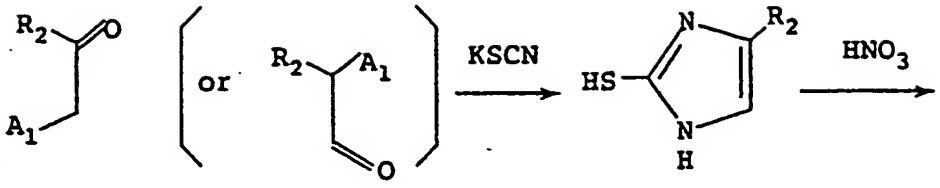

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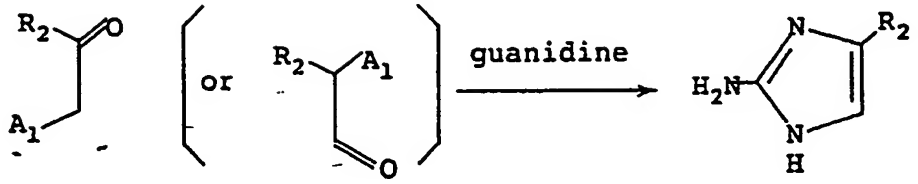
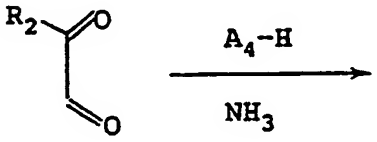


(i) diazotization/(ii) reduction

(31)



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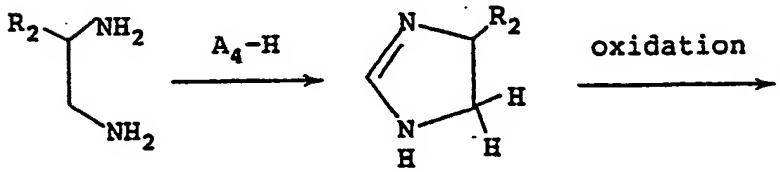

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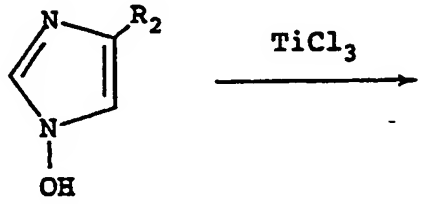


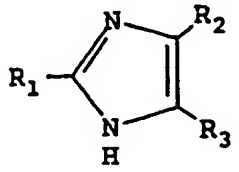
In the foregoing formulae, R_2 has the same meanings as described above; A_1 is a halogen atom, an amino group, a hydroxyl group, or an alkanoyloxy group; A_2 is a $-CONH_2$ group, a $-C(NH)NH_2$ group, or a $-C(NH)-A_3$ group, wherein A_3 is an alkoxy group or an alkylthio group; and A_4 is a formyl group.

The carbonyl group included in the above described formulae may be in the latent form of, for example, acetal, thioacetal, cyclic acetal, cyclic thioacetal, etc. Further, the formyl group represented by A_4 may be in the latent form of, for example, acetal, hemiacetal, etc.

In each of the processes as described above, the reaction conditions such as reaction temperature, reaction time, solvent, acid acceptor, alkali acceptor, etc. can appropriately be chosen from the conventionally known reaction conditions.

Typical examples of the intermediate compounds represented by the general formula (II), for the imidazole compounds of the present invention represented by the general formula (I) are shown in Table 1.

Table 1



(II)

Intermediate No.	R_1	R_2	R_3	Melting Point (°C)
1	CN	Br	H	196-201
2	"	3-trifluoromethyl-phenyl	"	160-168
3	"	Cl	CH_3	194-196
4	"	4-methoxyphenyl	Cl	150-155
5	"	phenyl	CH_3	222-225
6	"	"	Br	120-125
7	"	4-fluorophenyl	H	211-213
8	"	4-methylphenyl	"	228-232
9	"	"	Br	142-144
10	"	4-fluorophenyl	"	176-178
11	"	3,4-dichlorophenyl	H	115-121
12	"	4-methylphenyl	Cl	124-129
13	"	Cl	H	150-153
14	"	$n-C_3H_7$	Cl	107-109
15	"	phenyl	"	149-151

Table 1 (cont'd)

Intermediate No.	R_1	R_2	R_3	Melting Point (°C)
16	CN	3-methylphenyl	Cl	140-142
17	"	3,4-dimethylphenyl	"	150-152
18	"	4-fluorophenyl	"	153-155
19	"	4-bromophenyl	"	162-167
20	"	4-ethylphenyl	"	141-145
21	"	"	H	214-217
22	"	3-methoxyphenyl	"	218-220
23	"	4-nitrophenyl	"	230-235
24	"	5-chloro-2-thienyl	"	202-206
25	"	SCH_3	"	
26	"	phenylthio	"	166-169
27	"	phenyl	CN	207-215
28	"	H	F	
29	"	2-naphthyl	Cl	146-149
30	"	"	H	253-255
31	"	4-nitrophenyl	Cl	189-191
32	"	4-chlorophenyl	H	215-224
33	"	4-chlorophenyl	Cl	178-181
34	"	2-chlorophenyl	"	145-152
35	"	"	Br	152-156
36	"	4-isopropylphenyl	H	180-184
37	"	4-methylthiophenyl	"	217-219

Table 1 (cont'd)

Intermediate No.	R_1	R_2	R_3	Melting Point (°C)
38	CN	4-(2',2',2'-trifluoro-ethoxy)phenyl	H	195-198
39	"	CH_3	NO_2	125-130
40	"	tert-C_4H_9	Br	120-127
41	"	2-methylphenyl	H	
42	"	"	Cl	
43	"	5-methyl-2-furyl	H	169-171
44	"	3,4-dimethoxyphenyl	"	188-190
45	"	4-ethoxyphenyl	"	218-219
46	"	3-methyl-4-methoxy-phenyl	"	199-205
47	"	2-thienyl	"	195-203
48	"	4-(2',2',2'-trifluro-ethoxy)phenyl	Cl	164-166
49	"	"	Br	150-155
50	"	3-methyl-4-methoxy-phenyl	Cl	145-149
51	"	3-chloro-4-methyl-phenyl	Br	190-194
52	"	CH_3	CN	142-145
53	"	C_2H_5	H	127-129
54	"	"	Cl	138-140
55	"	n-C_3H_7	H	52-54
56	"	"	I	106-109
57	"	n-C_4H_9	H	83-85

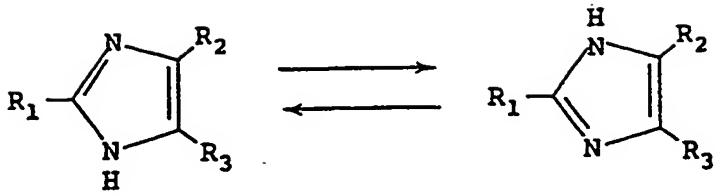
Table 1 (cont'd)

Intermediate No.	R_1	R_2	R_3	Melting Point (°C)
58	CN	n-C_4H_9	Cl	107-109
59	"	n-C_5H_{11}	H	89-92
60	"	n-C_5H_{11}	Cl	109-110
61	"	iso-C_3H_7	H	88-91
62	"	"	Cl	84-87
63	"	iso-C_4H_9	H	
64	"	"	Cl	142-145
65	"	tert-C_4H_9	H	130-135
66	"	"	Cl	120-124
67	"	iso-C_5H_{11}	H	144-146
68	"	"	Cl	104-107
69	"	cyclopropyl	"	170-183
70	"	cyclohexyl	H	185-190
71	"	"	Cl	130-133
72	"	3-chloropropyl	"	117-120
73	"	CH_2OCH_3	"	
74	"	$CH_2OC_2H_5$	"	
75	"	benzyl	"	144-146
76	"	phenethyl	"	147-152
77	"	SC_2H_5	H	112-115
78	"	"	Cl	128-131
79	"	S-n-C_4H_9	H	97-99
80	"	"	Cl	95-99

Table 1 (cont'd)

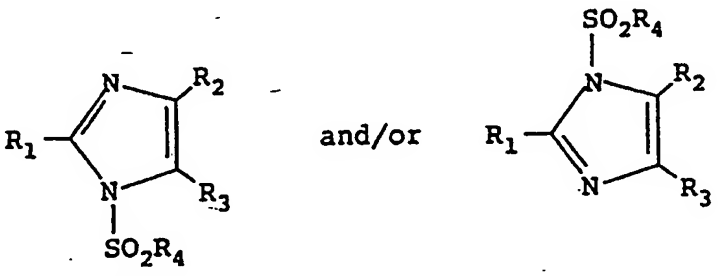
Intermediate No.	R_1	R_2	R_3	Melting Point (°C)
81	CN	3-fluoropropyl	Cl	
82	"	$SO_2N(CH_3)_2$	H	175-180
83	"	3-chlorophenyl	"	140-143
84	"	"	Cl	124-128
85	"	2,3-dichlorophenyl	H	202-206
86	"	"	Cl	198-204
87	"	3-chloro-4-methoxy-phenyl	"	158-160
88	"	"	Br	161-163
89	"	3-chloro-4-methyl-phenyl	Cl	165-169
90	"	4-cyanophenyl	H	240-244
91	"	"	Cl	250-255
92	"	"	Br	239-244
93	"	4-ethoxyphenyl	Cl	151-153
94	"	"	Br	140-145
95	"	2-fluorophenyl	H	190-195
96	"	"	Cl	155-159
97	"	2-methoxyphenyl	H	155-159
98	"	"	Cl	223-230
99	"	3,4-methylenedioxy-phenyl	H	228-231
100	"	"	Cl	149-152
101	"	"	Br	166-169

In the case that R_2 and R_3 are different from each other, the intermediate compounds represented by the general formula (II) described above include tautomers represented by the general formulae (II-a) and (II-b) described below:



(II-a) (II-b)

wherein R_1, R_2, and R_3 have the same meanings as described hereinabove. Accordingly, in the case that the imidazole compounds of the present invention represented by the general formula (I) are prepared using the compounds represented by the general formula (II) as a starting material, the imidazole compounds represented by the general formulae (I-a) and/or (I-b) described below can be obtained.



(I-a) and/or (I-b)

wherein R_1, R_2, R_3, and R_4 have the same meanings as described hereinabove. In the case that R_2 and R_3 are different from each other, the imidazole compounds represented by the general formulae (I-a) and (I-b) are tautomers each other. The same also applies to the compounds represented by the general formulae (I-1), (I-5), (I-6), and (I-7) in the processes [B] to [D] described hereinabove, etc.

The imidazole compounds represented by the general formula (I-a) or (I-b) described hereinabove can be separated concretely, for example, by methods [E-1] to [E-3] described below:

[E-1] Method by means of chromatography:

Each compound can be separated from a mixture of isomers of the general formulae (I-a) and (I-b) described above, by means of silica gel column chromatography, preparative high performance liquid chromatography, flash chromatography, etc. In the case of silica gel column chromatography, for example, n-hexane, carbon tetrachloride, methylene chloride, chloroform, ethyl acetate, or a mixture thereof can be used as a developing solvent.

[E-2] Method by means of recrystallization:

Each compound can be separated from a mixture of isomers of the general formulae (I-a) and (I-b) described above, using as a solvent for recrystallization, for example, carbon tetrachloride, methylene chloride, chloroform, 1,2-dichloroethane, ethyl acetate, diethyl ether, tetrahydrofuran, acetone, or a mixture thereof.

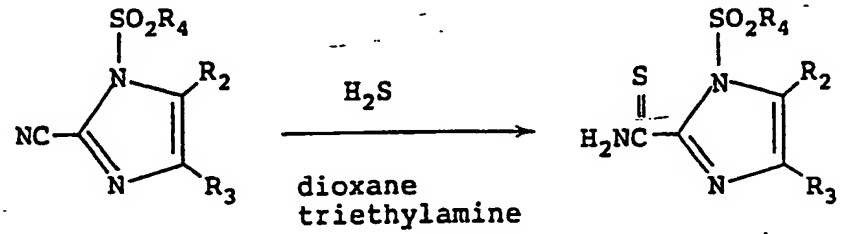
[E-3] Method by means of decomposition:

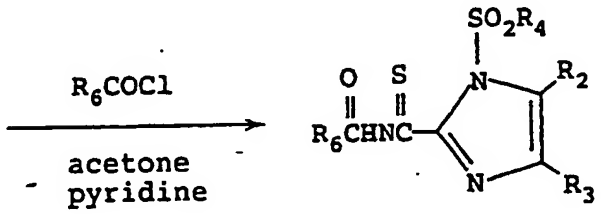
Either compound can be separated from a mixture of isomers of the general formulae (I-a) and (I-b) described above, by the selective hydrolysis under conditions of from 0 to 80 °C (preferably from room temperature to 50 °C) for from 1 to 48 hours (preferably from 5 to 24 hours).

As the mixture of isomers used in the methods [E-1] to [E-3] described above, it is preferred to use the mixture having a mixing ratio of both isomers as large as possible by appropriately choosing reaction conditions previously in the process [A] described above, for example, kind of solvent and acid acceptor and amounts thereof to be used, reaction temperature, reaction time, etc.

Further, in the case of preparing imidazole compounds wherein R_1 is a $-CSNH_2$ group or a $-CSNHR_5$ group, wherein R_5 has the same meaning as described hereinabove from compounds wherein R_1 is a cyano group in the compounds represented by the general formula (I-b) separated by the method [E-I], [E-2], or [E-3] described above, such compounds can be obtained, for example, by the following method:

[F]





wherein R_2, R_3, R_4, and R_6, have the same meanings as described hereinabove.

Specific examples of synthesizing the imidazole compounds of the present invention are described below.

Synthesis Example 1

Synthesis of 2-cyano-1-dimethylsulfamoylimidazole (Compound No. 1)

Thirty grams of 2-cyanoimidazole, 53.4 g of anhydrous potassium carbonate and 600 ml of acetonitrile were mixed at room temperature. After reacting for 2 hours at the refluxing temperature, the reaction mixture was cooled, and 55.6 g of dimethylsulfamoyl chloride was added thereto. The mixture was reacted again at the refluxing temperature for 2 hours.

After completion of the reaction, the reaction mixture was poured into water. Extraction with methylene chloride was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation. The obtained residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 28.0 g of 2-cyano-1-dimethylsulfamoylimidazole (Compound No. 1) having a melting point of from 74 to 76 °C.

Synthesis Example 2

Synthesis of 2-cyano-1-dimethylsulfamoyl-5-phenylthioimidazole (Compound No. 10-b)

In a four-necked flask were charged 12.0 g of 2-cyano-1-dimethylsulfamoylimidazole (Compound No. 1) and 240 ml of dry tetrahydrofuran in a nitrogen flow. While maintaining the mixture at -75° C or below with dry ice-acetone, 41.3 ml of a 1.6 M n-butyl lithium hexane solution (manufactured by Aldrich) was gradually added dropwise to the mixture. After completion of the dropwise addition, the system was kept at the same temperature for 15 minutes. Then, a solution of 17 g of diphenyl disulfide in 30 ml of tetrahydrofuran was added dropwise to the mixture at -70° C or below. While stirring overnight, the temperature was gradually reverted to room temperature.

After completion of the reaction, the reaction mixture was poured into water. Extraction with 500 ml of ethyl acetate was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The ethyl acetate was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 4.3 g of 2-cyano-1-dimethylsulfamoyl-5-phenylthioimidazole (Compound No. 10-b) having a melting point of from 106 to 107° C.

Synthesis Example 3

Synthesis of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole (Compound No. 16-b)

[1] 4.8 g of 2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole having a melting point of from 51 to 52° C (Compound No. 3-b) was synthesized by the reaction of 12.0 g of 2-cyano-1-dimethylsulfamoylimidazole (Compound No. 1) and 15.3 g of n-propyl iodide in a manner similar to Synthesis Example 2 described above.

[2] 4.8 g of 2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole as obtained in [1] above, 40 ml of pyridine, and 11.4 g of pyridinium chloride were mixed, and the mixture was stirred at 90° C for 4 hours. After completion of the reaction, the pyridine was removed by distillation from the reaction mixture, and the residue was extracted with ethyl acetate. The extract was washed with water and then dried over anhydrous sodium sulfate. Thereafter, the ethyl acetate was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: a mixture of ethyl acetate and n-hexane) and separated to give 2.46 g of 2- cyano-4(5)-n-propylimidazole (Intermediate No. 55) having a melting point of from 52 to 54° C.

[3] 2.35 g of 2-cyano-4(5)-n-propylimidazole as obtained in [2] above, 80 ml of chloroform, and 2.6 g of N-chlorosuccinimide were mixed, and the mixture was reacted at the refluxing temperature for 4 hours. After completion of the reaction, 200 ml of water was added to the reaction mixture. The resulting organic layer was washed with water and then dried over anhydrous sodium sulfate. After drying, the chloroform was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: a 1:1 mixture of ethyl acetate and n-hexane) and separated to give 2.2 g of 4(5)-chloro-2-cyano-5-(4)-n-propylimidazole (Intermediate No. 14) having a melting point of from 107 to 109° C.

[4] 2.0 g of 4(5)-chloro-2-cyano-5(4)-n-propylimidazole as obtained in [3] above, 30 ml of acetonitrile, 1.95 g of anhydrous potassium carbonate, and 1.86 g of dimethylsulfamoyl chloride were mixed, and after gradually elevating the temperature, the mixture was reacted at the refluxing temperature for 1 hours. After completion of the reaction, the acetonitrile was removed by distillation from the reaction mixture. After pouring 100 ml of water into the residue, the resulting mixture was extracted with 50 ml of methylene chloride. The extract was washed with water and dried over anhydrous sodium sulfate. Thereafter, the methylene chloride was removed by distillation. The residue was allowed to stand overnight, and the analysis thereof revealed that one of the two isomers in the mixture decomposed and returned to the starting 4(5)-chloro-2-cyano-5(4)-n-propylimidazole. The residue containing the other isomer was purified by silica gel column chromatography (developing solvent: methylene chloride) and separated to give 1.1 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole (Compound No. 16-b) having a melting point of from 64 to 66° C.

Synthesis Example 4

Synthesis of 2-cyano-1-dimethylsulfamoyl-4(5)-phenylimidazole (Compound No. 4)

[1] In 320 ml of acetone was dissolved 23.04 g of 4(5)-phenylimidazole, and 12.14 g of anhydrous potassium carbonate was added to the solution. The mixture was heated at the refluxing temperature for 2 hours. After cooling, 45 ml of an acetone solution containing 25.25 g of dimethylsulfamoyl chloride was added dropwise to the mixture. After completion of the dropwise addition, the mixture was heated at the refluxing temperature for 4.5 hours to complete the reaction.

After completion of the reaction, the reaction mixture was cooled, and solid substances were removed by filtration. After the solvent was removed by distillation under reduced pressure, the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 17.8 g of 1-dimethylsulfamoyl-4(5)-phenylimidazole having a melting point of from 96 to 100 °C.

[2] In 290 ml of tetrahydrofuran was dissolved 17 g of 1-dimethylsulfamoyl-4(5)-phenylimidazole as obtained in [1] above. The solution was cooled to -70 °C in a nitrogen flow, and 51 ml of a 1.6 M n-butyl lithium hexane solution was added dropwise to the mixture over 30 minutes. After completion of the dropwise addition, the reaction mixture was stirred at -70 °C for 30 minutes. Then, 12 ml of a tetrahydrofuran solution containing 6 g of N,N-dimethylformamide was added dropwise to the mixture. After completion of the dropwise addition, the reaction mixture was reacted for 15 hours with stirring while slowly elevating the temperature to room temperature.

After completion of the reaction, the reaction mixture was poured into ice water and extracted with ethyl acetate. After washing the extracted layer with water, the extracted layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: a 1:2 mixture of ethyl acetate and n-hexane) to give 12.8 g of 1- dimethylsulfamoyl-2-formyl-4(5)-phenylimidazole having a melting point of from 86 to 89 °C.

[3] In 120 ml of pyridine were dissolved 11.16 g of 1-dimethylsulfamoyl-2-formyl-4(5)-phenylimidazole as obtained in [2] above and 5.56 g of hydroxylamine hydrochloride, and 24 ml of acetic anhydride was added dropwise to the solution at room temperature. After completion of the dropwise addition, the temperature was gradually raised, and the mixture was reacted at 100 °C for 12 hours.

After completion of the reaction, the solvent in the reaction mixture was removed by distillation under reduced pressure. Then, 125 ml of water was added to the residue, and the precipitated solid was separated by filtration. The crude product was dissolved in ethyl acetate and purified by silica gel column chromatography (developing solvent: ethyl acetate) to give 5.55 g of 2-cyano-4(5)-phenylimidazole having a melting point of from 203 to 205 °C.

[4] In 88 ml of acetone was dissolved 1.7 g of 2-cyano-4(5)-phenylimidazole as obtained in [3] above, and 1.7 g of anhydrous potassium carbonate was added to the solution. The mixture was heated at the refluxing temperature for 2 hours.

After cooling, 6 ml of an acetone solution containing 1.7 g of dimethylsulfamoyl chloride was added dropwise to the mixture. After completion of the dropwise addition, the mixture was heated at the refluxing temperature for 2 hours to complete the reaction.

After completion of the reaction, the reaction mixture was cooled, and solid substances were removed by filtration. After the solvent was removed by distillation under reduced pressure, the residue was extracted with ethyl acetate. The extract was washed with water and dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure to give 2 g of 2-cyano-1-dimethylsulfamoyl-4(5)-phenylimidazole (Compound No. 4) having a melting point of from 101 to 102 °C.

Synthesis Example 5

Synthesis of 4(5)-chloro-2-cyano-1-dimethylsulfamoyl-5(4)-phenylimidazole (Compound No. 17) and 4-chloro-2-cyano-1-dimethylsulfamoyl-5-phenylimidazole (Compound No. 17-b)

[1] In 100 ml of chloroform was dissolved 1.352 g of 2-cyano-4(5)-phenylimidazole, and 1.175 g of N-chlorosuccinimide was added to the solution. The mixture was reacted upon heating at the refluxing temperature for 4 hours.

After completion of the reaction, the reaction mixture was poured into water and extracted with chloroform. After washing with water, the extracted layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1.28 g of 4(5)-chloro-2-cyano-5(4)-phenylimidazole (Intermediate No. 15) having a melting point of from 149 to 151 °C.

[2] In 6 ml of acetone was dissolved 0.43 g of 4(5)-chloro-2-cyano-5(4)-phenylimidazole as obtained in [1] above, and 0.29 g of anhydrous potassium carbonate and 0.36 g of dimethylsulfamoyl chloride were added to the solution. The mixture was reacted upon heating at the refluxing temperature for 30 minutes.

After completion of the reaction, the reaction mixture was poured into water and extracted with ethyl acetate. After washing with water, the extracted layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was then purified by silica gel column chromatography (developing solvent: methylene chloride) to give 0.5 g of 4(5)-chloro-2-cyano-1-dimethylsulfamoyl-5(4)-phenylimidazole (Compound No. 17) having a melting point of from 106 to 109 °C.

As a result of analysis by means of NMR spectra, the compound described above was an isomer mixture of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-phenylimidazole and 5-chloro-2-cyano-1-dimethylsulfamoyl-4-phenylimidazole in almost equal ratios.

[3] After allowing to stand for 24 hours at room temperature, 2.9 g of the mixture of these isomers as obtained in a manner similar to [2] above was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1.15 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-phenylimidazole (Compound No. 17-b) having a melting point of from 109 to 112 °C. Further, by purification of and isolation from this compound, 0.7 g of 4(5)-chloro-2-cyano-5(4)-phenylimidazole (Intermediate No. 15) was also obtained.

Synthesis Example 6

Synthesis of 4(5)-chloro-2-cyano-1-dimethylsulfamoyl-5(4)-(4-methylphenyl)imidazole (Compound No. 18) and 4-chloro-2-cyano-1-dimethylsulfamoyl-5-(4-methylphenyl)imidazole (Compound No. 18-b)

An isomer mixture (Compound No. 18), having a melting point of from 101 to 108 °C, of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-(4-methylphenyl)imidazole and 5-chloro-2-cyano-1-dimethylsulfamoyl-4-(4-methylphenyl)imidazole was obtained from 4(5)-(4-methylphenyl)imidazole in a ratio of 6:4 in a manner similar to Synthesis Examples 4 and 5 described above. After 0.75 g of the isomer mixture was reacted at 40 °C for 8 hours, the reaction mixture was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 0.45 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-(4-methylphenyl)-imidazole (Compound No. 18-b) having a melting point of from 133 to 134 °C. Further, by purification of and isolation from this compound, 0.15 g of 4(5)-chloro-2-cyano-5(4)-(4-methylphenyl)imidazole (Intermediate No. 12) having a melting point of from 124 to 129 °C was also obtained.

Synthesis Example 7

Synthesis of 4(5)-chloro-5(4)-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 23), 4-chloro-5-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 23-b) and 5-chloro4-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 23-a)

In a manner similar to Synthesis Examples 4 and 5 described above, 0.80 g of an isomer mixture (Compound No. 23), having a melting point of 108 °C, of 4-chloro-5-(4-chlorophenyl)-2-cyano-1-dimethylsul-

famoylimidazole and 5-chloro-4-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole was obtained from 4(5)-(4-chlorophenyl)imidazole. The isomer mixture was purified by silica gel column chromatography (developing solvent: methylene chloride). The eluate of the second fraction was concentrated and recrystallized from methylene chloride to give 0.16 g of 4-chloro-5-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 23-b) having a melting point of from 117 to 120 °C. Further, the eluate of the first fraction was likewise concentrated and recrystallized from methylene chloride to give 0.50 g of 5-chloro-4-(4-chlorophenyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 23-a) having a melting point of from 133 to 138 °C.

Synthesis Example 8

Synthesis of 1-dimethylsulfamoyl-4(5)-phenylimidazole-2-carbothioamide (Compound No. 49)

In 30 ml of dioxane was dissolved 1.0 g of 2-cyano-1-dimethylsulfamoyl-4(5)-phenylimidazole (Compound No. 4), and 0.36 g of triethylamine was added to the solution. The mixture was heated to 40 to 50 °C while stirring, and a hydrogen sulfide gas was introduced thereinto for one hour and 25 minutes. Thereafter, the mixture was reacted at 40 to 50 °C for an additional 50 minutes.

After completion of the reaction, the reaction mixture was cooled, poured into water, and extracted with ethyl acetate. After washing with water, the extracted layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: a 1:3 mixture of ethyl acetate and n-hexane) to give 0.8 g of 1-dimethylsulfamoyl-4(5)-phenyl-imidazole-2-carbothioamide (Compound No. 49) having a melting point of from 155 to 175 °C. Crystals of 4(5)-phenylimidazole-2-carbothioamide were also obtained in a small quantity.

Synthesis Example 9

Synthesis of 2-cyano-1-isopropylsulfonyl-4(5)-phenylimidazole (Compound No. 101)

One gram of 2-cyano-4(5)-phenylimidazole, 0.98 g of anhydrous potassium carbonate, and 30 ml of acetonitrile were mixed at room temperature. After reacting for 2 hours at the refluxing temperature, the reaction mixture was cooled, and a solution of 1.0 g of isopropylsulfonyl chloride in 5 ml of acetonitrile was added thereto. The mixture was reacted again at the refluxing temperature for 1.5 hours.

After completion of the reaction, the reaction mixture was poured into water. Extraction with methylene chloride was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1.4 g of 2-cyano-1-isopropylsulfonyl-4(5)-phenylimidazole (Compound No. 101) having a melting point of from 80 to 83 °C.

Synthesis Example 10

Synthesis of 4(5)-(2-thienyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 6)

[1] To 150 ml of formamide was added 25 g of 2-(bromoacetyl)thiophene. The mixture was reacted at 180 to 190 °C for 2 hours.

After completion of the reaction, the reaction mixture was poured into water, and concentrated hydrochloric acid was added thereto to render the system acidic. Then, washing with methylene chloride

was carried out. After neutralizing with ammonia water, the aqueous phase was extracted with methylene chloride. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure to give 11 g of 4(5)-(2-thienyl)imidazole.

[2] To 200 ml of acetonitrile were added 11.6 g of dimethylsulfamoyl chloride, 11.1 g of anhydrous potassium carbonate, and 11 g of 4(5)-(2-thienyl)imidazole as obtained in [1] above. The mixture was reacted for 2 hours while stirring.

After completion of the reaction, the reaction mixture was poured into water. Extraction with ethyl acetate was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure to give 14.5 g of 4(5)-(2-thienyl)-1-dimethylsulfamoylimidazole.

[3] In 120 ml of anhydrous tetrahydrofuran was dissolved 9.5 g of 4(5)-(2-thienyl)-1-dimethylsulfamoylimidazole as obtained in [2] above. In a nitrogen flow, 26.2 ml of a 1.6 M n-butyl lithium hexane solution was added dropwise to the solution at -78° C, and the mixture was stirred at the same temperature for 15 minutes. Then, 20 ml of a tetrahydrofuran solution having dissolved therein 5.4 g of N,N-dimethylformamide was added dropwise to the mixture. After completion of the dropwise addition, the temperature was gradually reverted to room temperature to complete the reaction.

After completion of the reaction, the reaction mixture was poured into water. Extraction with ethyl acetate was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure to give 5.4 g of 4(5)-(2-thienyl)-2-formyl-1-dimethylsulfamoylimidazole.

[4] In 54 ml of pyridine were dissolved 2.6 g of hydroxylamine hydrochloride and 5.4 g of 4(5)-(2-thienyl)-2-formyl-1-dimethylsulfamoylimiazole as obtained in [3] above. The solution was stirred at room temperature for 15 minutes. Then, 10 ml of acetic anhydride was gradually added to the solution, followed by reacting at 60 to 70° C for 2 hours.

After completion of the reaction, the reaction mixture was poured into water. Extraction with ethyl acetate was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: a 2:1 mixture of ethyl acetate and n-hexane) to give 1.2 g of 4(5)-(2- thienyl) 2-cyanoimiazole (Intermediate No. 47) having a melting point of from 195 to 203° C.

[5] To 50 ml of acetonitrile were added 1.1 g of dimethylsulfamoyl chloride, 1.0 g of anhydrous potassium carbonate, and 1.2 g of 4(5)-(2-thienyl)-2-cyanoimidazole as obtained in [4] above. The mixture was reacted at the refluxing temperature for 2 hours.

After completion of the reaction, the reaction mixture was poured into water and extracted with ethyl acetate. After drying the extract over anhydrous sodium sulfate, the solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1.3 g of 4(5)-(2-thienyl)-2-cyano-1-dimethylsulfamoylimidazole (Compound No. 6) having a melting point of from 145 to 150° C

Synthesis Example 11

Synthesis of 4(5)-chloro-2-cyano-1-dimethylsulfamoyl-5(4)-isopropylimidazole (Compound No. 125) and 4-chloro-2-cyano-1-dimethylsulfamoyl-5-isopropylimidazole (Compound No. 125-b)

[1] 360 g of formamide was heated to 180° C, and 102 g of 1-hydroxy-3-methyl-2-butanone (prepared in a manner as described in Lipshutz and Morey, J. Org. Chem., 48, 3745 (1983)) was added dropwise thereto over 30 minutes. After completion of the dropwise addition, the mixture was reacted at 180° C for one hour.

After completion of the reaction, the reaction mixture was cooled and poured into ice water. The resulting mixture was adjusted at a pH of 1 with hydrochloric acid and washed with methylene chloride. The aqueous layer was adjusted at a pH of 4 to 5 with ammonia water. 5 g of activated charcoal was added thereto, and the mixture was stirred for one hour. The activated charcoal was removed by filtration, and the filtrate was adjusted at a pH of 8 with ammonia water. Then, extraction with methylene chloride was carried out, and the extract was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure to give 13 g of 4(5)-isopropylimidazole.

[2] In 300 ml of acetonitrile was dissolved 11.8 g of 4(5)-isopropylimidazole as obtained in [1] above, and 18 g of anhydrous potassium carbonate was added to the solution. The mixture was refluxed for 30 minutes, and after cooling, 17 g of dimethylsulfamoyl chloride was added dropwise thereto. After completion of the dropwise addition, the mixture was refluxed to complete the reaction.

After completion of the reaction, the reaction mixture was cooled, poured into water, and then extracted with ethyl acetate. The extracted layer was washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 13 g of 1-dimethylsulfamoyl-4(5)-isopropylimidazole.

[3] In 200 ml of tetrahydrofuran was dissolved 13 g of 1-dimethylsulfamoyl-4(5)-isopropylimidazole as obtained in [2] above. The solution was cooled to -70°C in a nitrogen flow, and 38 ml of a 1.6 M n-butyl lithium hexane solution was added dropwise thereto over 15 minutes. After completion of the dropwise addition, the mixture was stirred at -70°C for 30 minutes. After dropwise addition of 5.6 g of N,N-dimethylformamide, the mixture was reacted with stirring for 15 hours while slowly elevating the temperature to room temperature.

After completion of the reaction, the reaction mixture was poured into ice water and extracted with ethyl acetate. The extracted layer was washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure to obtain 8.6 g of 1-dimethylsulfamoyl-2-formyl-4(5)-isopropylimidazole.

[4] In 100 ml of pyridine were dissolved 8.5 g of 1-dimethylsulfamoyl-2-formyl-4(5)-isopropylimidazole as obtained in [3] above and 4.8 g of hydroxylamine hydrochloride, and 10 ml of acetic anhydride was added dropwise to the solution at room temperature. After completion of the dropwise addition, the temperature was gradually elevated, and the mixture was reacted at 80 to 90°C for 5 hours.

After completion of the reaction, the solvent in the reaction mixture was distilled off under reduced pressure. To the residue was added water, and the mixture was extracted with ethyl acetate. The extracted layer was washed with dilute hydrochloric acid and then with water and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure to give 2.35 g of 2-cyano-4(5)-isopropylimidazole (Intermediate No. 61) having a melting point of from 88 to 91°C.

[5] In 80 ml of methanol was dissolved 2 g of 2-cyano-4(5)-isopropylimidazole as obtained in [4] above, and 2.1 g of N-chlorosuccinimide was added to the solution. The mixture was stirred at room temperature for 20 hours and then reacted at 40°C for 8 hours.

After completion of the reaction, the methanol in the reaction mixture was distilled off under reduced pressure. To the residue was added water, and the mixture was extracted with ethyl acetate. The extracted layer was washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1.67 g of 4(5)-chloro-2-cyano-5(4)-isopropylimidazole (Intermediate No. 62) having a melting point of from 84 to 87°C.

[6] In 30 ml of acetonitrile was dissolved 1.6 g of 4(5)-chloro-2-cyano-5(4)-isopropylimidazole as obtained in [5] above, and 1.56 g of anhydrous potassium carbonate was added to the solution. The mixture was refluxed for 30 minutes. After cooling, 1.49 g of dimethylsulfamoyl chloride was added dropwise thereto. After completion of the dropwise addition, the mixture was refluxed for 15 minutes to complete the reaction.

After completion of the reaction, the reaction mixture was cooled, poured into water, and then extracted with ethyl acetate. The extracted layer was washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 2.1 g of 4(5)-chloro-2-cyano-1-dimethylsulfamoyl-5(4)-isopropylimidazole (Compound No. 125).

As a result of analysis by means of NMR spectra, the compound described above was an isomer mixture of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-isopropylimidazole and 5-chloro-2-cyano-1-dimethylsulfamoyl-4-isopropylimidazole in a proportion of about 2:1.

[7] After allowing to stand for 5 days at room temperature, 2.1 g of the isomer mixture as obtained in [6] above was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 1 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-isopropylimidazole (Compound No. 125-b) having a melting point of from 75 to 82°C (decomposed). Further, by purification of and isolation from this compound, 4(5)-chloro-2-cyano-5(4)-isopropylimidazole (Intermediate No. 62) was also obtained.

Synthesis Example 12

Synthesis of-4-chloro-1-dimethylsulfamoyl-5-n-propylimidazole-2-carbothioamide (Compound No. 185-b)

[1] In a four-necked flask were charged 6.0 g of 2-cyano-4,5-dichloro-1-dimethylsulfamoylimidazole having a melting point of from 100 to 103 °C and 180 ml of dry tetrahydrofuran in a nitrogen flow. While maintaining the mixture at -75 °C or below with dry ice-acetone, 15.3 ml of a 1.6 M n-butyl lithium hexane solution (manufactured by Aldrich) was gradually added dropwise to the mixture. After completion of the dropwise addition, the system was kept at the same temperature for 15 minutes. Then, a solution of 5.7 g of n-propyl iodide in 15 ml of tetrahydrofuran was added dropwise to the mixture at -70 °C or below. While stirring overnight, the temperature was gradually reverted to room temperature.

After completion of the reaction, the reaction mixture was poured into water. Extraction with 500 ml of methylene chloride was carried out. After washing with water, the extract was dried over anhydrous sodium sulfate. The methylene chloride was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) and then again purified by silica gel column chromatography (developing solvent: a mixture of ethyl acetate and n-hexane) to give 2.8 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole (Compound No. 16-b) having a melting point of from 66 to 68 °C.

[2] In a four-necked flask were charged 2.7 g of 4-chloro-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole as obtained in [1] above, 40 ml of dioxane, 1.0 g of triethylamine, and 0.8 g of pyridine. Into this mixture was introduced a hydrogen sulfide gas at 20 to 25 °C for about 30 minutes until the starting materials had disappeared.

After completion of the reaction, the reaction mixture was poured into water, and precipitated crystals were filtered by means of a Nutsche and dried. The resulting crystals were purified by silica gel column chromatography (developing solvent: methylene chloride) and separated to give 2.3 g of 4-chloro-1-dimethylsulfamoyl-5-n-propylimidazole-2-carbothioamide (Compound No. 185-b) having a melting point of from 160 to 162 °C.

Synthesis Example 13

Synthesis of N-propionyl-4-chloro-1-dimethylsulfamoyl-5-n-propylimidazole-2-carbothioamide (Compound No. 187-b

Into a four-necked flask were charged 2.0 g of 4-chloro-1-dimethylsulfamoyl-5-n-propylimidazole-2-carbothioamide (Compound No. 185-b), 24 ml of acetone, and 1.12 g of pyridine. 1.19 g of propionyl chloride was added dropwise to the mixture at 0 to 5 °C. After completion of the dropwise addition, the reaction was carried out at 30 to 35 °C for one hour and at the refluxing temperature for an additional 30 minutes with stirring.

After completion of the reaction, the reaction mixture was poured into water and extracted with ethyl acetate. The extracted layer was washed with water and dried over anhydrous sodium sulfate. Thereafter, the ethyl acetate was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) and separated to give 1.02 g of N-propionyl-4-chloro-1-dimethylsulfamoyl-5-n-propylimidazole-2-carbothioamide (Compound No. 187-b) having a melting point of from 150 to 152 °C.

Synthesis Example 14

Synthesis of 2-cyano-1-dimethylsulfamoyl-4,5-diphenylthioimidazole (Compound No. 141)

[1] 8.0 g of 2-cyano-1-dimethylsulfamoyl-5-phenylthioimidazole (Compound No. 10-b) as obtained in a similar manner to Synthesis Example 2 described above, 60 ml of methanol, and 60 ml of a 7% hydrochloric acid aqueous solution were charged, and the mixture was reacted with stirring at 40 to 50 °C

for 2 hours. After completion of the reaction, the reaction mixture was rendered weakly alkaline with ammonia, and precipitated crystals were separated by filtration and dried to give 4.2 g of 2-cyano-4(5)-phenylthioimidazole (Intermediate No. 26) having a melting point of from 166 to 169 °C.

[2] To a mixture of 4.2 g of 2-cyano-4(5)-phenylthioimidazole as obtained in [1] above, 80 ml of acetonitrile, and 3.1 g of anhydrous potassium carbonate. was added 3.4 g of dimethylsulfamoyl chloride. The resulting mixture was reacted at the refluxing temperature for one hour. After completion of the reaction, the reaction mixture was cooled, and solid substances were filtered. The solvent in the filtrate was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) and separated to give 5.8 g of 2-cyano-1-dimethylsulfamoyl-4(5)-phenylthioimidazole (Compound No. 10.

[3] In a four-necked flask were charged 5.8 g of 2-cyano-1-dimethylsulfamoyl-4(5)-phenylthioimidazole as obtained in [2] above and 150 ml of dry tetrahydrofuran in a nitrogen atmosphere, and 12.9 ml of a 1.6 M n-butyl lithium hexane solution (manufactured by Kanto Kagaku) was added dropwise to the mixture while maintaining the temperature at -75 °C or below with dry ice-acetone. After completion of the dropwise addition, the mixture was kept at the same temperature for 15 minutes, and 20 ml of a solution of 5.2 g of diphenyl disulfide in tetrahydrofuran was added dropwise thereto at -70 °C or below. Thereafter, the mixture was returned to room temperature. After completion of the reaction, the reaction mixture was extracted with ethyl acetate. The extract was washed with water and dried over anhydrous sodium sulfate. The solvent was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) and separated to give 1.7 g of 2-cyano-1-dimethylsulfamoyl-4,5-diphenylthioimidazole (Compound No. 141) having a melting point of from 98 to 101 °C.

Synthesis Example 15

Synthesis of 4-bromo-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole (Compound No. 157-b)

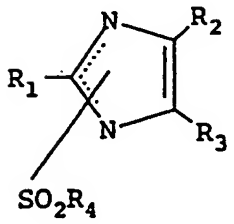
[1] 2-Cyano-4,5-dibromo-1-dimethylsulfamoylimidazole having a melting point of from 118 to 120 °C was synthesized from 2-cyano-4,5-dibromoimidazole having a melting point of from 200 to 203 °C in a similar manner to Synthesis Example 1 described above.

[2] In a 200 ml four-necked flask were charged 5 g of 2-cyano-4,5-dibromo-1-dimethylsulfamoylimidazole as obtained in [1] above and 120 ml of dry tetrahydrofuran in a nitrogen flow. While maintaining the mixture at -75 °C or below with dry ice-acetone, 9.6 ml of a 1.6 M n-butyl lithium hexane solution (manufactured by Aldrich) was gradually added dropwise to the mixture. After completion of the dropwise addition, the system was kept at the same temperature for 15 minutes. Then, a solution of 3.6 g of n-propyl iodide in 15 ml of tetrahydrofuran was added dropwise to the mixture at -75 °C or below. While stirring, the temperature was gradually reverted to room temperature.

After completion of the reaction, the reaction mixture was extracted with ethyl acetate. After washing with water, the extract was dried over anhydrous sodium sulfate. The ethyl acetate was removed by distillation, and the residue was purified by silica gel column chromatography (developing solvent: methylene chloride) to give 2.1 g of 4-bromo-2-cyano-1-dimethylsulfamoyl-5-n-propylimidazole (Compound No. 157-b) having a melting point of from 93 to 94 °C.

Typical examples of the imidazole compounds (general formula (I)) of the present invention are shown in Table 2.

Table 2



(I)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
1	CN	H	H	$-N(CH_3)_2$	74-76
2	"	CH_3	"	"	78-83
3	"	$n-C_3H_7$	"	"	
4	"	phenyl	"	"	101-102
5	"	4-chlorophenyl	"	"	148-149
6	"	2-thienyl	"	"	145-150
7	"	5-chloro-2-thienyl	"	"	145-148
8	"	5-bromo-2-thienyl	"	"	138-140
9	"	SCH_3	"	"	
10	"	phenylthio	"	"	
11	"	2-chlorophenyl	"	"	118-121
12	"	4-nitrophenyl	"	"	107-108
13	"	4-trifluoromethylphenyl	"	"	
14	"	H	Cl	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
15	CN	CH_3	Cl	$-N(CH_3)_2$	
16	"	n-C_3H_7	"	"	
17	"	phenyl	"	"	106-109
18	"	4-methylphenyl	"	"	101-108
19	"	3-methylphenyl	"	"	90-95
20	"	2-methylphenyl	"	"	
21	"	3,4-dimethyl-phenyl	"	"	95-105
22	"	4-methoxyphenyl	"	"	102-107
23	"	4-chlorophenyl	"	"	108
24	"	2-chlorophenyl	"	"	
25	"	3,4-dichloro-phenyl	"	"	99-105
26	"	4-fluorophenyl	"	"	105-107
27	"	SCH_3	"	"	
28	"	phenylthio	"	"	
29	"	H	Br	"	
30	"	CH_3	"	"	
31	"	tert-C_4H_9	"	"	88-90
32	"	phenyl	"	"	
33	"	4-methylphenyl	"	"	106-108
34	"	4-tert-butyl-phenyl	"	"	105-110
35	"	4-methoxyphenyl	"	"	96-99

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
36	CN	4-fluorophenyl	Br	$-N(CH_3)_2$	87-93
37	"	4-chlorophenyl	"	"	
38	"	1,2-dibromo-ethyl	Cl	"	
39	"	C_2H_5	Br	"	
40	"	$-CH_2CH=CH_2$	"	"	
41	"	4-bromophenyl	Cl	"	110-116
42	"	4-isopropyl-phenyl	"	"	
43	"	2-naphthyl	"		124-126
44	"	CH_3	CH_3	"	52-54
45	"	phenyl	"	"	101-105
46	"	"	SCH_3	"	
47	"	"	phenyl	"	148-149
48	"	"	CN	"	124-129
49	$-CSNH_2$	phenyl	H	"	155-175
50	"	4-chlorophenyl	"	"	197-201
51	"	phenyl	Cl	"	110-130
52	"	H	Br	"	140-144
53	"	phenyl	"	"	
54	CN	3,4-dimethoxy-phenyl	H	"	
55	"	3-methyl-4-methoxyphenyl	Cl	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
56	CN	4-ethylphenyl	Cl	$-N(CH_3)_2$	
57	"	phenylthio	Br	"	
58	"	benzyl	"	"	
59	"	3-chloropropyl	H	"	
60	"	$-SO_2C_2H_5$	"	"	
61	"	3-fluoropropyl	Cl	"	
62	"	4-methylthio-phenyl	H	"	
63	"	vinyl	Cl	"	
64	"	5-methyl-2-thienyl	H	"	
65	"	2-chlorophenyl	Br	"	
66	"	3,4-dichloro-phenyl	H	"	139-142
67	"	4-(2',2',2'-trifluoroethoxy)-phenyl	Cl	"	
68	"	"	Br	"	
69	"	$-CH_2OH$	H	"	
70	"	3-chlorophenyl	Cl	"	
71	"	3-fluorophenyl	"	"	
72	"	2-fluorophenyl	"	"	96-101
73	"	$-SCH_2CH=CH_2$	H	"	
74	"	CH_3	NO_2	"	110-117

Table 2 (cont'd)

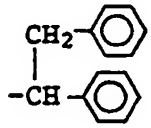
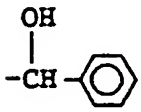
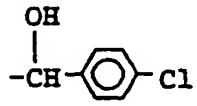
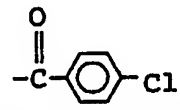
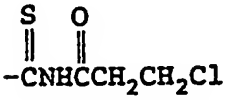
Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
75	CN	 (1,2-diphenylethyl)	H	$-N(CH_3)_2$	
76	"	 (α-hydroxybenzyl)	"	"	
77	"	 (4-chloro-α-hydroxybenzyl)	"	"	
78	"	 (4-chlorobenzoyl)	"	"	
79	"	acetyl	"	"	
80	 (N-acetylthiocarbamoyl)	phenyl	"	"	
81	 [N-(3-chloropropionyl)-thiocarbamoyl]	"	"	"	

Table 2 (cont'd)

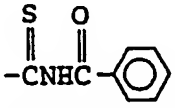
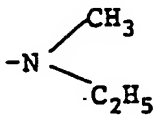
Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
82	N-acetyl-thiocarbamoyl	CH_3	H	$-N(CH_3)_2$	
83	 (N-benzoylthiocarbamoyl)	"	"	"	
84	CN	5-methyl-2-furyl	"	"	120-124
85	"	C_2H_5	Cl	1-piperidinyl	
86	"	"	"	phenyl	
87	"	4-(chloromethylthio)-phenyl	H	$-N(CH_3)_2$	142-146
88	"	CH_3	CN	"	80-84
89	"	cyclohexyl	H	"	
90	"	$-SO_2CH_3$	"	"	
91	"	4-chlorobenzenesulfonyl	"	"	
92	"	phenyl	Cl	C_2H_5	
93	"	"	"	cyclohexyl	
94	"	"	"	CF_3	
95	"	"	"	2-thienyl	
96	"	"	"		

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
97	CN	phenyl	Cl	$-N(CH_3)(CH_2CF_3)$	
98	"	"	"	$-N(CH_3)(CH_2CH=CH_2)$	
99	"	"	"	1-pyrrolidinyl	
100	"	4-methylphenyl	"	morpholino	
101	"	phenyl	H	isopropyl	80-83
102	"	"	Cl	"	
103	"	"	"	$-N(C_2H_5)_2$	70-80
104	"	"	Br	"	55-76
105	"	"	Cl	morpholino	106-110
106	"	"	Br	"	70-83
107	"	"	Cl	thiomorpholino	
108	"	4-(2'-chloroethyl)phenyl	"	$-N(CH_3)_2$	
109	"	4-chlorobenzyl	Br	"	
110	"	benzyl	H	"	
111	"	4-chlorophenylthio	Cl	"	
112	"	3-chloropropyl	"	"	
113	"	C_2H_5	"	"	
114	"	2-furyl	2-furyl	"	118-123
115	"	4-pyridyl	H	"	138-142

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
116	CN	2-thienyl	Cl	$-N(CH_3)_2$	
117	"	4-fluoro-n-butyl	"	"	
118	"	5-fluoropentyl	"	"	
119	"	$n-C_4H_9$	"	"	
120	"	$n-C_5H_{11}$	"	"	
121	"	$n-C_6H_{13}$	"	"	
122	"	$n-C_7H_{15}$	"	"	
123	"	$n-C_8H_{17}$	"	"	
124	"	$n-C_{12}H_{25}$	"	"	
125	"	$iso-C_3H_7$	"	"	
126	"	$iso-C_4H_9$	"	"	
127	"	$tert-C_4H_9$	"	"	
128	"	cyclopropyl			
129	"	cyclohexyl	"	"	
130	"	$-CH_2CH=CH_2$	"	"	
131	"	geranyl ($C_{10}H_{17}$)	"	"	
132	"	SC_2H_5	"	"	
133	"	$S-n-C_3H_7$	"	"	
134	"	$S-n-C_4H_9$	"	"	36-38
135	"	benzylthio	"	"	
136	"	3-trifluoro-methyl-2-pyridylthio	"	"	
137	"	"	H	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
138	CN	4-chlorophenyl-thio	H	$-N(CH_3)_2$	
139	"	$S\text{-}n\text{-}C_3H_7$	"	"	
140	"	SC_2H_5	"	"	
141	"	phenylthio	phenylthio	"	98-101
142	"	"	C_2H_5	"	
143	"	benzene-sulfonyl	H	"	
144	"	2-fluoro-benzenesulfon-yl	"	"	
145	"	4-chlorobutyl	Cl	"	
146	"	5-chloropentyl	"	"	
147	"	CH_2OCH_3	"	"	
148	"	$CH_2OC_2H_5$	"	"	
149	"	1-hydroxypropyl	"	"	
150	"	1-hydroxybutyl	"	"	
151	"	benzyl	"	"	94-97
152	"	4-methylbenzyl	"	"	
153	"	3-methylbenzyl	"	"	
154	"	2-methylbenzyl	"	"	
155	"	2-fluorobenzyl	"	"	
156	"	phenethyl	"	"	
157	"	$n\text{-}C_3H_7$	Br	"	
158	"	$n\text{-}C_4H_9$	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
159	CN	$n\text{-}C_5H_{11}$	Br	$-N(CH_3)_2$	
160	"	$n\text{-}C_6H_{13}$	"	"	
161	"	$iso\text{-}C_3H_7$	"	"	
162	"	$iso\text{-}C_4H_9$	"	"	
163	"	cyclopropyl	"	"	
164	"	cyclohexyl	"	"	
165	"	4-chloro-phenylthio	"	"	
166	"	OCH_2CF_3	"	"	77-79
167	"	$S\text{-}n\text{-}C_3H_7$	"	"	
168	"	$S\text{-}n\text{-}C_4H_9$	"	"	
169	"	$S\text{-}iso\text{-}C_4H_9$	"	"	
170	"	CH_2OCH_3	"	"	
171	"	$CH_2OC_2H_5$	"	"	
172	"	methoxycarbonyl	"	"	
173	"	N-(4-chloro-phenyl)carbamoyl	"	"	
174	"	N-phenyl-carbamoyl	"	"	
175	"	N-ethyl-carbamoyl	"	"	
176	$-CSNH_2$	C_2H_5	Cl	"	
177	N-acetyl-thiocarba-moyl	"	"	"	
178	$-CSNH_2$	$n\text{-}C_4H_9$	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
179	N-acetyl-thiocarbamoyl	$n-C_4H_9$	Cl	$-N(CH_3)_2$	
180	CN	H	I	"	101-105
181	"	$n-C_3H_7$	"	"	
182	"	"	$-COCF_3$	"	
183	$-CSNH_2$	"	Br	"	
184	N-acetyl-thio-carbamoyl	"	"	"	
185	$-CSNH_2$	"	Cl	"	
186	N-acetyl-thiocarbamoyl	"	"	"	
187	N-propionylthio-carbamoyl	"	"	"	
188	N-methyl-thio-carbamoyl	phenyl	"	"	
189	N-acetyl-thio-carbamoyl	"	"	"	
190	CN	$-SO_2N(CH_3)_2$	H	"	142-149
191	"	$-Si(CH_3)_3$	Cl	"	
192	"	$n-C_{10}H_{21}$	"	"	
193	"	C_2H_5	H	"	
194	"	$n-C_4H_9$	"	"	
195	"	$S-n-C_4H_9$	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
196	CN	1-hydroxy-3-phenylpropyl	Cl	$-N(CH_3)_2$	
197	"	1-hydroxypropyl	H	"	
198	"	α-hydroxybenzyl	Cl	"	
199	"	α-acetoxybenzyl	"	"	
200	"	1-hydroxy-3-methylbutyl	"	"	
201	"	4-methyl-3-chlorophenyl	"	"	
202	"	"	Br	"	
203	"	4-methoxy-3-chlorophenyl	Cl	"	
204	"	"	Br	"	
205	"	2,3-dichloro-phenyl	Cl	"	
206	"	4-ethoxyphenyl	"	"	
207	"	"	Br	"	
208	"	3,4-methylene-dioxyphenyl	Cl	"	
209	"	"	Br	"	
210	"	4-cyanophenyl	Cl	"	
211	"	"	Br	"	
212	"	4-nitrophenyl	Cl	"	140-145
213	"	2-butenyl	"	"	
214	"	iso-C_5H_{11}	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
215	$-CSNH_2$	H	Cl	$-N(CH_3)_2$	
216	"	CH_3	"	"	
217	"	C_5H_{11}	"	"	
218	"	benzyl	"	"	
219	N-acetylthio-carbamoyl	H	"	"	
220	"	CH_3	"	"	
221	"	C_5H_{11}	"	"	
222	"	benzyl	"	"	
223	N-propionyl-thiocabamoyl	"	"	"	
224	$-CSNH_2$	C_2H_5	Br	"	
225	N-acetylthio-carbamoyl	"	"	"	
226	N-propionyl-thiocarbamoyl	"	"	"	
227	CN	3-chlorobutyl	Cl	"	
228	"	$-CF_2CF=CF_2$	H	"	
229	"	sec-C_4H_9	Cl	"	
230	"	$-CH_2CH=C(CH_3)_2$	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
3-b	CN	$n\text{-}C_3H_7$	H	$-N(CH_3)_2$	51-52
9-b	"	SCH_3	"	"	114-115
10-b	"	phenylthio	"	"	106-107
14-b	"	H	Cl	"	111-114
15-b	"	CH_3	"	"	90-95
16-b	"	$n\text{-}C_3H_7$	"	"	64-66
17-b	"	phenyl	"	"	109-112
18-b	"	4-methylphenyl	"	"	133-134
19-b	"	3-methylphenyl	"	"	
20-b	"	2-methylphenyl	"	"	93-96
21-b	"	3,4-dimethyl-phenyl	"	"	
22-b	"	4-methoxyphenyl	"	"	
23-a	"	4-chlorophenyl	"	"	133-138
23-b	"	"	"	"	117-120
24-b	"	2-chlorophenyl	"	"	113-117
25-b	"	3,4-dichloro-phenyl	"	"	
26-b	"	4-fluorophenyl	"	"	120-122
27-b	"	SCH_3	"	"	101-103
28-b	"	phenylthio	"	"	107-108
29-b	"	H	Br	"	100-103
30-b	"	CH_3	"	"	107-110
31-b	"	$tert\text{-}C_4H_9$	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
32-b	CN	phenyl	Br	$-N(CH_3)_2$	122-124
33-b	"	4-methylphenyl	"	"	136-137
34-b	"	4-tert-butyl-phenyl	"	"	
35-b	"	4-methoxyphenyl	"	"	
36-b	"	4-fluorophenyl	"	"	
37-b	"	4-chlorophenyl	"	"	
39-b	"	C_2H_5	"	"	112-115
40-b	"	$-CH_2CH=CH_2$	"	"	92-94
41-b	"	4-bromophenyl	Cl	"	
42-a	"	4-isopropyl-phenyl	"	"	110-114
42-b	"	"	"	"	135-138
43-b	"	2-naphthyl	"	"	
46-b	"	phenyl	SCH_3	"	99-101
49-b	$-CSNH_2$	"	H		
50-b	"	4-chlorophenyl	"	"	
51-b	"	phenyl	Cl	"	115-128
52-b	"	H	Br	"	
53-b	"	phenyl	"	"	110-118
55-b	CN	3-methyl-4-methoxyphenyl	Cl	"	115-118
56-b	"	4-ethylphenyl	"	"	110-112
57-b	"	phenylthio	Br	"	94-97

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
58-b	CN	benzyl	Br	$-N(CH_3)_2$	87-89
59-b	"	3-chloropropyl	H	"	
60-b	"	$-SO_2C_2H_5$	"	"	121-124
61-b	"	3-fluoropropyl	Cl	"	75-79
65-b	"	2-chlorophenyl	Br	"	119-123
67-b	"	4-(2',2',2'-trifluoro-ethoxy)phenyl	Cl	"	111-113
68-b	"	"	Br	"	115-118
69-b	"	$-CH_2OH$	H	"	106-107
70-b	"	3-chlorophenyl	Cl	"	96-99
71-b	"	3-fluorophenyl	"	"	
72-b	"	2-fluorophenyl	"	"	
73-b	"	$-SCH_2CH=CH_2$	H	"	20-30
75-b	"	1,2-diphenyl-ethyl	"	"	101-103
76-b	"	α-hydroxybenzyl	"	"	98-100
103-b	"	phenyl	Cl	$-N(C_2H_5)_2$	99-101
104-b	"	"	Br	"	
105-b	"	"	Cl	morpholino	
106-b	"	"	Br	"	126-130
111-b	"	4-chlorophenyl-thio	Cl	$-N(CH_3)_2$	92-94
112-b	"	3-chloropropyl	"	"	102-105
113-b	"	C_2H_5	"	"	95-97
119-b	"	n-C_4H_9	"	"	48-49

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
120-b	CN	$n\text{-}C_5H_{11}$	Cl	$-N(CH_3)_2$	37-39
121-b	"	$n\text{-}C_6H_{13}$	"	"	$n_D^{23.5}$ 1.5002
122-b	"	$n\text{-}C_7H_{15}$	"	"	$n_D^{23.5}$ 1.5019
123-b	"	$n\text{-}C_8H_{17}$	"	"	$n_D^{23.6}$ 1.4981
124-b	"	$n\text{-}C_{12}H_{25}$	"	"	34-36
125-b	"	$iso\text{-}C_3H_7$	"	"	75-82 (decomposed)
126-b	"	$iso\text{-}C_4H_9$	"	"	73-76
127-b	"	$tert\text{-}C_4H_9$	"	"	74-80
128-b	"	cyclopropyl	"	"	76-79
129-b	"	cyclohexyl	"	"	107-111
130-b	"	$-CH_2CH=CH_2$	"	"	67-72
131-b	"	geranyl ($C_{10}H_{17}$)	"	"	
132-b	"	SC_2H_5	"	"	107-110
133-b	"	$S\text{-}n\text{-}C_3H_7$	"	"	70-74
134-b	"	$S\text{-}n\text{-}C_4H_9$	"	"	
135-b	"	benzylthio	"	"	149-152
136-b	"	3-trifluoromethyl-2-pyridylthio	"	"	126-127
137-b	"	"	H	"	109-111
138-b	"	4-chlorophenylthio	"	"	110-112
140-a	"	SC_2H_5	"	"	36-40

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
140-b	CN	SC_2H_5	H	$-N(CH_3)_2$	41-45
142-a	"	phenylthio	C_2H_5	"	86-89
145-b	"	4-chlorobutyl	Cl	"	$n_D^{22.1}$ 1.5382
146-b	"	5-chloropentyl	"	"	$n_D^{24.8}$ 1.5328
147-b	"	CH_2OCH_3	"	"	64-66
148-b	"	$CH_2OC_2H_5$	"	"	82-84
149-b	"	1-hydroxypropyl	"	"	70-73
150-b	"	1-hydroxybutyl	"	"	$n_D^{24.2}$ 1.5097
151-b	"	benzyl	"	"	92-100
152-b	"	4-methylbenzyl	"	"	125-129
153-b	"	3-methylbenzyl	"	"	93-96
154-b	"	2-methylbenzyl	"	"	119-123
155-b	"	2-fluorobenzyl	"	"	105-109
156-b	"	phenethyl	"	"	106-110
157-b	"	$n-C_3H_7$	Br	"	93-94
158-b	"	$n-C_4H_9$	"	"	
159-b	"	$n-C_5H_{11}$	"	"	
160-b	"	$n-C_6H_{13}$	"	"	99-101
161-b	"	$iso-C_3H_7$	"	"	
162-b	"	$iso-C_4H_9$	"	"	
163-b	"	cyclopropyl	"	"	
164-b	"	cyclohexyl	"	"	

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
165-b	CN	4-chlorophenyl-thio	Br	$-N(CH_3)_2$	94-95
167-b	"	S-n-C_3H_7	"	"	76-78
168-b	"	S-n-C_4H_9	"	"	48-50
169-b	"	S-iso-C_4H_9	"	"	77-79
170-b	"	CH_2OCH_3	"	"	65-67
171-b	"	$CH_2OC_2H_5$	"	"	100-101
172-b	"	methoxycarbonyl	"	"	98-101
173-b	"	N-(4-chloro-phenyl)carbamoyl	"	"	106-109
174-b	"	N-phenyl-carbamoyl	"	"	105-107
175-b	"	N-ethyl-carbamoyl	"	"	98-101
181-a	"	n-C_3H_7	I	"	76-79
181-b	"	"	"	"	99-103
182-a	"	"	$-COCF_3$	"	90-92
185-b	$-CSNH_2$	"	Cl	"	160-162
186-b	N-acetyl-thio-carbamoyl	"	"	"	119-123
187-b	N-propion-ylthio-carbamoyl	"	"	"	150-152
188-b	N-methyl-thio-carbamoyl	phenyl	"	"	67-72

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
189-b	N-acetyl-thio-carbamoyl	phenyl	Cl	$-N(CH_3)_2$	110-114
191-b	CN	$-Si(CH_3)_3$	"	"	116-119
192-b	"	$n-C_{10}H_{21}$	"	"	$n_D^{23.6}$ 1.4994
193-b	"	C_2H_5	H	"	69-71
194-b	"	$n-C_4H_9$	"	"	52-53
195-b	"	$S-n-C_4H_9$	"	"	50-51
196-b	"	1-hydroxy-3-phenylpropyl	Cl	"	$n_D^{24.0}$ 1.5512
197-b	"	1-hydroxypropyl	H	"	94-97
198-b	"	α-hydroxybenzyl	Cl	"	102-104
199-b	"	α-acetoxybenzyl	"	"	82-86
200-b	"	1-hydroxy-3-methylbutyl	"	"	71-74
201-b	"	4-methyl-3-chlorophenyl	"	"	99-103
202-b	"	"	Br	"	103-106
203-b	"	4-methoxy-3-chlorophenyl	Cl	"	97-101
204-b	"	"	Br	"	105-110
205-b	"	2,3-dichloro-phenyl	Cl	"	103-107
206-b	"	4-ethoxyphenyl	"	"	122-124
207-b	"	"	Br	"	110-113
208-b	"	3,4-methylene-dioxyphenyl	Cl	"	150-153

Table 2 (cont'd)

Compound No.	R_1	R_2	R_3	R_4	Melting Point (°C)
209-b	CN	3,4-methylene-dioxyphenyl	Br	$-N(CH_3)_2$	95-98
210-b	"	4-cyanophenyl	Cl	"	182-185
211-b	"	"	Br	"	175-178
212-b	"	4-nitrophenyl	Cl	"	144-146
213-b	"	2-butenyl	"	"	87-90
214-b	"	iso-C_5H_{11}	"	"	45-47
218-b	$-CSNH_2$	benzyl	"	"	118-121
222-b	N-acetyl-thiocarbamoyl	"	"	"	163-165
223-b	N-propionyl-thiocarbamoyl	"	"	"	149-152
227-b	CN	3-chlorobutyl	"	"	54-57
230-b	"	$-CH_2CH=C(CH_3)_2$	"	"	75-78

Among the imidazole compounds of the present invention described in Table 2 above, the compounds having a mark "a" in their compound numbers are ones falling within the general formula (I-a) in the general formula (I) described hereinabove and the compounds having a mark "b" in their compound numbers are ones falling within the general formula (I-b) in the general formula (I) described hereinabove.

The imidazole compounds of the present invention are useful as biocides for controlling harmful organisms in the agricultural, horticultural, medical, and pharmaceutical areas.

As agricultural and horticultural fungicides, the compounds exhibit an excellent effect of controlling diseases of crop plants such as rice blast caused by Pyricularia oryzae, rice sheath blight caused by Rhizoctonia solani, oat crown rust caused by Puccinia coronata, cucumber anthracnose caused by Colletotrichum lagenarium, cucumber powdery mildew caused by Sphaerotheca fuliginea, cucumber downy mildew caused by Pseudoperonospora cubensis, tomato late blight caused by Phytophthora infestans, tomato early blight caused by Alternaria solani, citrus melanose caused by Diaporthe citri citrus common green mold caused by Penicillium digitatum, pear scab caused by Venturia nashicola, apple alternaria blotch caused by Alternaria mali, grape downy mildew caused by Plasmopara viticola, and further gray mold caused by Botrytis cinerea and sclerotinia rot caused by Sclerotinia sclerotiorum of various crops, etc.; or soil diseases caused by phytopathogenic fungi such as Fusarium, Pythium, Rhizoctonia, Verticillium, Plasmodiophora, Aphanomyces, etc.

In particular, the compounds exhibit an excellent effect of preventing deseases such as potato or tomato late blight caused by Phytophthora infestans, cucumber downy mildew caused by Pseudoperonospora cubensis, grape downy mildew caused by Plasmopara viticola, and tobacco blue mold caused by Peronospora tabacina; and soil diseases caused by phycomycetes such as Plasmodiophora, Aphanomyces, Pythium, etc.

The compounds of the present invention have a prolonged residual effect so that they exhibit an excellent preventing effect, and further exhibit an excellent curative effect as well. Therefore, it is possible to

control deseases by treatment after infection. The compounds of the present invention are appropriate to be applied to crop plants by foliar treatment. Further, the compounds possess a systemic activity so that it is also possible to control deseases of the stem and leaf by soil treatment. In addition, the compounds of the present invention show an excellent controlling effect against agriculturally and horticulturally harmful insects such as various planthoppers, diamondback moth (Plutella xylostella), green rice leafhopper (Nephotettix cincticeps), adzuki bean weevil (Callosobruchus chinensis), common cutworm (Spodoptera litura), green peach aphid (Myzus persicae), etc.; mites such as two-spotted spider mite (Tetranychus urticae), carmine spider mite (Tetranychus cinnabarinus), citrus red mite (Panonychus citri), etc.; and nematodes such as southern root-knot nematode (Meloidogyne incognita), etc.

Upon use, the compounds of the present invention can be prepared into a variety of forms of biocidal compositions such as emulsifiable concentrates, suspension concentrates, dusts, wettable powders, aqueous solutions, granules, etc., together with adjuvants, as in conventional formulations. Upon actual use of these formulations, they can be used as such or by diluting with a diluent such as water or the like to a predetermined concentration.

As the adjuvants used herein, mention may be made of carriers, emulsifying agents, suspending agents, dispersing agents, spreaders, penetrating agents, wetting agents, thickeners, stabilizers, etc.

The carriers are classified into solid carriers and liquid carriers. As the solid carriers, mention may be made of animal and vegetable powders such as starch, sugar, cellulose powders, cyclodextrin, activated charcoal, soybean powders, wheat powders, chaff-powders, wood powders, fish powders, powdery milk, etc.; and mineral powders such as talc, kaolin, bentonite, bentonite-alkylamine complex, calcium carbonate, calcium sulfate, sodium bicarbonate, zeolite, diatomaceous earth, white carbon, clay, alumina, silica, sulfur powders, etc. As the liquid carriers, mention may be made of water; animal and vegetable oils such as corn oil, soybean oil, cotton seed oil, etc.; alcohols such as ethyl alcohol, ethylene glycol, etc.; ketones such as acetone, methyl ethyl ketone, etc.; ethers such as dioxane, tetrahydrofuran, etc.; aliphatic hydrocarbons such as kerosene, lamp oil, liquid paraffin, etc.; aromatic hydrocarbons such as xylene, trimethylbenzene, tetramethylbenzene, cyclohexane, solvent naphtha, etc.; halogenated hydrocarbons such as chloroform, chlorobenzene, etc.; acid amides such as dimethylformamide, etc.; esters such as ethyl acetate, fatty acid glycerine esters, etc.; nitriles such as acetonitrile, etc.; sulfur-containing compounds such as dimethyl sulfoxide, etc.; and N-methyl pyrrolidone, etc.

The adjuvants other than the carriers described hereinabove, such as emulsifying agents, suspending agents, dispersing agents, spreaders, penetrating agents, wetting agents, thickeners, stabilizers, etc. are exemplified more specifically as following surfactants.

Polyoxyethylene alkylarylether, polyoxyethylene glycol nonyl phenylether, polyoxyethylene laurylether, polyoxyethylene caster oil, polyoxyethylene alkylaryl sulfate (polyoxyethylene alkylphenyl ether sulfate), polyoxyethylene fatty acid ester (polyoxyethylene stearate), polyoxyethylene sorbitan fatty acid ester, lower alcohol phosphate, sodium alkylsulfate, sodium lignin sulfonate, calcium lignin sulfonate, alkylaryl sulfonate, sodium alkylbenzene sulfonate, sodium β-naphthalene sulfonate-formaldehyde condensate, dialkylsulfosuccinate.

The compound of the present invention is uniformly mixed with at least one kind of adjuvants described hereinabove to form a biocidal composition.

A weight ratio of the compound of the present invention to the adjuvants to be formulated is generally from 0.05:99.95 to 90:10, preferably from 0.2:99.8 to 80:20.

Since a concentration of the compound of the present invention to be applied may vary depending upon crop to be applied, method for application, preparation form, dose to be applied, etc., it is difficult to define a specific concentration range. However, if it is forced to define specifically, the concentration of the compound is generally from 0.1 to 10,000 ppm, desirably from 1 to 2,000 ppm in the case of foliar treatment, and is generally from 10 to 100,000 g/ha, desirably from 200 to 20,000 g/ha in the case of soil treatment.

Further, if necessary and desired, the compound of the present invention can be used as admixture with or in combination with other agricultural chemicals, for example, insecticides, acaricides, nematocides, fungicides, antiviral agents, attractants, herbicides, plant growth regulators, etc. In this case, more excellent effects can sometimes be exhibited.

As the insecticides, acaricides or nematocides, mention may be made of, for example, organic phosphrous compounds, carbamate compounds, organic chlorine compounds, organic metal compounds, pyrethroid compounds, benzoyl urea compounds, juvenile hormone-like compounds, dinitro compounds, organic sulfur compounds, urea compounds, triazine compounds, etc. The compound of the present invention can also be used as admixture with or in combination with biological pesticides such as BT agents, insect pathogenic viral agents, etc.

As the fungicides, mention may be made of, for example, organic phosphorus compounds, organic chlorine compounds, dithiocarbamate compounds, N-halogenothioalkyl compounds, dicarboximide compounds, benzimidazole compounds, azole compounds, carbinol compounds, benzanilide compounds, acylalanine compounds, pyridinamine compounds, piperazine compounds, morpholine compounds, anthraquinone compounds, quinoxaline compounds, crotonic acid compounds, sulfenic acid compounds, urea compounds, antibiotics, etc.

On the other hand, as medical and pharmaceutical antimicrobial agent, the compounds of the present invention are effective against microorganisms belonging to Staphylococcus and Trichophyton.

Upon use, the compounds can be orally and unorally administered similarly to the conventional medicines.

In the case of oral administating use, the compounds may be formulated into various types suited for gastroenteral absorption such as tablets, granules, capsules, syrup, aqueous or oily suspensions, and the like.

And, in the case of unoral administrating use, compounds may be formulated for injection or into various types suited for cuteneous absorption such as creams, ointments, and the like.

Preferable dose varies according to the conditions such as etat, age, etc. of human beings and animals infected with pathogen.

Hereafter, test examples of the biocidal compositions for controlling harmful organisms in the agricultural, horticultural, medical, and pharmaceutical areas in accordance with the present invention are described below.

Standards for evaluation of the agricultural and horticultural fungicides follow the following criteria for evaluation, unless otherwise indicated.

Standards for Evaluation

The controlling effect was determined by visually observing a degree of desease of a test plant and expressed by the following 5 grades of the index of control.

[Index of Control]	[Degree of Desease]
5:	No lesion is noted at all.

4: Area, number or length of lesions is less than 10% as compared to the non-treated plot.

3: Area, number or length of lesions is less than 40% as compared to the non-treated plot.

2: Area, number or length of lesions is less than 70% as compared to the non-treated plot.

1: Area, number or length of lesions is more than 70% as compared to the non-treated plot.

Test Example 1

Test on preventive effect against cucumber powdery mildew

Cucumber (cultivars: Suyo) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When cucumber reached the one-leaf stage, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over cucumber using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C over one day and one night, conidia of fungi of powdery mildew (Sphaerotheca fuliginea) were inoculated. Ten days after the inoculation, an area of lesion on the first leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 3 were obtained.

Table 3

Compound No.	Index of Control 500 ppm
15-b	4
23-a	4
59-b	4
106-b	3
133-b	4
167-b	3
169-b	3
171-b	5

Test Example 2

Test on preventive effect against cucumber anthracnose

Cucumber (cultivars: Suyo) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When cucumber reached the two-leaf stage, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over cucumber using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24°C over one day and one night, a spore suspension of fungi of anthracnose (Colletotrichum lagenarium) was inoculated. Seven days after the inoculation, an area of lesion on the first leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 4 were obtained. .

Table 4

Compound No.	Index of Control 500 ppm
3-b	3
17-b	3
26	5
28-b	3
51	3
51-b	3
59-b	3
69-b	3
70-b	4
73-b	3
75-b	3
101	4
105	4
106	3

Test Example 3

Test on preventive effect against cucumber downy mildew

Cucumber (cultivars: Suyo) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When cucumber reached the two-leaf stage, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over cucumber using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C over one day and one night, a spore suspension of fungi of downy mildew (Pseudoperonospora cubensis) was inoculated. Six days after the inoculation, an area of lesion on the first leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 5 were obtained.

Table 5

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
4	5	5	29-b	5	5
5	5	5	30-b	5	5
6	5	4	31	4	3
7	5	5	32-b	5	5
8	5	3	33	-	5
14-b	5	3	34	5	5
15-b	5	5	36	-	5
16-b	-	5	37	5	5
17	5	5	45	5	5
17-b	-	5	47	5	5
23	5	5	48	5	5

Table 5 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
49	5	4	101	5	4
50	5	5	103	5	5
52	5	5	105	4	-
53-b	5	5	106	5	5

Test Example 4

Test on curative effect against cucumber downy mildew

Cucumber (cultivars: Suyo) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When cucumber reached the two-leaf stage, a spore suspension of fungi of downy mildew (Pseudoperonospora cubensis) was inoculated. Six hours after the inoculation, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over cucumber using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C for 6 days, an area of lesion on the first leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 6 were obtained.

Table 6

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
3-b	5	-	8	5	-

Table 6(cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
9-b	5	-	32-b	5	-
10-b	5	-	33	5	-
12	5	-	33-b	5	-
14-b	5	-	36	5	-
15-b	5	-	37	5	-
16-b	5	-	39-b	-	5
17	5	-	40-b	-	5
17-b	5	-	41	-	5
18	-	5	46-a	5	-
18-b	5	-	48	4	-
19	5	-	51	5	-
20-b	5	-	51-b	5	-
22	5	-	52	5	-
23	5	-	53-b	5	-
23-a	5	-	56-b	5	-
23-b	5	-	57-b	-	5
24-b	-	5	58-b	-	5
25	-	4	59-b	-	5
26	-	5	60-b	-	5
26-b	5	-	61-b	-	5
27-b	5	-	65-b	5	-
28-b	5	-	67-b	5	-
29-b	5	-	68-b	-	4
30-b	5	-	69-b	4	-

Table 6 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
70-b	5	-	138-b	-	5
72	-	5	141	4	-
74	-	4	142-a	5	5
76-b	5	-	145-b	-	5
88	-	5	146-b	-	5
101	4	-	147-b	5	5
103-b	5	-	148-b	5	5
106-b	5	-	149-b	-	5
111-b	-	5	150-b	5	-
112-b	5	5	151	-	5
113-b	5	5	151-b	5	5
119-b	5	5	152-b	-	3
120-b	5	5	153-b	-	5
121-b	5	5	154-b	-	5
125-b	-	5	155-b	-	5
126-b	-	5	156-b	-	5
128-b	-	5	157-b	-	5
129-b	-	5	160-b	5	5
130-b	5	5	166	5	3
132-b	-	5	167-b	5	5
133-b	5	4	169-b	5	5
134	5	5	170-b	5	5
135-b	4	-	171-b	5	5
136-b	-	3	173-b	4	-

Table 6 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	Compound No.	Index of Control 125 ppm	31 ppm
180	5	-	201-b	4	-
181-a	-	5	203-b	3	-
181-b	-	5	208-b	4	3
185-b	-	5	209-b	5	-
186-b	-	5	210-b	4	-
187-b	-	5	212-b	5	5
189-b	5	5	213-b	-	5
190	5	4	214-b	-	5

Test Example 5

Test on systemic effect against cucumber downy mildew

Cucumber (cultivars: Suyo) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When cucumber reached the two-leaf stage, 15 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was drenched on the surface of soil using a pipette. After keeping the pots in a constant temperature chamber of 22 to 24° C for 2 days, a spore suspension of fungi of downy mildew (Pseudoperonospora cubensis) was inoculated. Six days after the inoculation, an area of lesion on the first leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 7 were obtained.

Table 7

Compound No.	Index of Control 500 ppm	125 ppm
1	5	3
14-b	5	5
15-b	-	5
17	5	4
29-b	5	5
30-b	5	5
37	5	5
52	5	5
53-b	5	5

Test Example 6

Test on preventive effect against tomato late blight

Tomato (cultivars: Ponderosa) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When tomato reached the four-leaf stage, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over tomato using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C over one day and one night, a zoosporangium suspension of fungi of late blight (Phytophthora infestans) was inoculated. Five days after the inoculation, an area of lesion on the leaves was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 8 were obtained.

Table 8

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
3-b	-	5	-
4	5	4	-
5	5	5	-
6	5	4	-
7	5	5	-
8	5	5	-
9-b	-	5	-
10-b	-	-	5
12	-	5	3
14-b	-	5	-
15-b	-	4	-
16-b	-	5	4
17	5	5	-
17-b	5	5	-
18	-	5	-
18-b	-	5	-
19	-	5	-
20-b	-	5	-
21	-	5	-

Table 8 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
22	-	5	-
23	5	5	-
23-a	-	5	-
23-b	-	5	-
24-b	-	5	5
25	-	5	5
26	-	5	-
26-b	-	5	-
27-b	-	5	-
28-b	-	5	-
29-b	5	5	-
30-b	-	5	-
32-b	5	5	-
33	5	5	-
33-b	-	5	-
34	4	4	-
36	5	5	-
37	-	5	-
39-b	-	-	5
40-b	-	5	5
41	-	5	5
42-a	-	5	-
42-b	-	5	-
43	-	-	5

Table 8 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
45	5	5	-
46-a	5	5	-
48	5	5	-
49	5	3	-
50	4	-	-
51	5	5	-
51-b	5	5	-
52	5	4	-
53-b	5	5	-
55-b	-	4	5
56-b	-	5	-
57-b	-	-	5
58-b	-	5	5
59-b	-	-	5
60-b	-	5	-
61-b	-	5	5
65-b	-	5	-
66	5	5	-
67-b	-	-	5
68-b	-	-	5
70-b	-	-	5
72	-	-	5
73-b	-	4	-
74	-	-	5

Table 8 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
75-b	-	5	-
76-b	-	5	-
84	-	-	5
88	-	-	5
101	5	5	-
103	5	-	-
104	5	4	-
105	5	4	-
106-b	5	4	-
111-b	-	-	4
112-b	-	5	5
113-b	-	-	5
114	-	5	5
119-b	-	5	5
120-b	-	5	5
121-b	-	5	5
122-b	-	5	5
123-b	-	5	5
124-b	-	5	5
125-b	-	-	5
126-b	-	-	5
128-b	-	5	4
129-b	-	5	5
130-b	-	5	5

Table 8 (cont'd)

Compound No.	Index of Control		
	125 ppm	31 ppm	8 ppm
132-b	-	5	5
133-b	-	-	5
134	-	5	5
135-b	-	5	5
136-b	-	-	5
137-b	-	-	5
138-b	-	-	4
141	-	5	5
142-a	-	5	5
145-b	-	-	4
146-b	-	5	5
147-b	-	4	3
148-b	-	4	-
149-b	-	-	5
151	-	-	5
151-b	-	5	5
152-b	-	-	5
153-b	-	-	5
154-b	-	-	5
155-b	-	-	5
156-b	-	-	5
157-b	-	-	5

Table 8 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
160-b	-	5	5
166	-	5	3
167-b	-	5	5
169-b	-	5	5
170-b	-	5	3
171-b	-	5	-
173-b	-	4	3
174-b	-	4	-
180	-	-	5
181-b	-	5	5
182-b	-	5	5
185-b	-	-	5
186-b	-	-	5
187-b	-	-	5
189-b	-	5	4
190	-	4	-
201-b	-	5	5
202-b	-	5	5
203-b	-	4	5
205-b	-	-	5
206-b	-	5	5
207-b	-	5	-
208-b	-	5	5

Table 8 (cont'd)

Compound No.	Index of Control 125 ppm	31 ppm	8 ppm
209-b	-	4	-
210-b	-	4	3
211-b	-	4	-
212-b	-	5	3
213-b	-	5	5
214-b	-	5	5

Test Example 7

Test on systemic effect against tomato late blight

Tomato (cultivars: Ponderosa) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When tomato reached the four-leaf stage, 15 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was drenched on the surface of soil using a pipette. After keeping the pots in a constant temperature chamber of 22 to 24° C for 2 days, a zoosporangium suspension of fungi of late blight (Phytophthora infestans) was inoculated. Five days after the inoculation, an area of lesion on the leaves was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 9 were obtained.

Table 9

Compound No.	Index of Control 500 ppm	125 ppm
3-b	-	4
10-b	5	5
16-b	-	4
17-b	5	4
19	4	4
20-b	5	4
22	5	4
27-b	5	5
28-b	5	-
40-b	5	5
51	5	5
51-b	5	5
57-b	-	4
58-b	5	3
59-b	-	4
76-b	-	5

Test Example 8

Test on preventive effect against rice blast

Rice plant (cultivars: Chukyo Asahi) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When rice plant reached the four-leaf stage, 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over rice plant using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C over one day and one night, a spore suspension of fungi of blast (Pyricularia oryzae) was inoculated. Five days after the inoculation, a number of lesion was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 10 were obtained.

Table 10

Compound No.	Index of Control 500 ppm
27-b	4
48	3
53-b	3
55-b	4
134	3
167-b	3
201-b	4
202-b	4

Test Example 9

Test on preventive effect against rice sheath blight

Rice plant (cultivars: Chukyo Asahi) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When rice plant reached the five-leaf stage, 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over rice plant using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24°C over one day and one night, rice straw in which fungi of sheath blight (Rhizoctonia solani) had been previously incubated was set between leaf sheath portions to inoculate. After keeping the pots in an inoculation room having a temperature of 28°C and a humidity of 100% for 5 days, a length of lesion was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 11 were obtained.

Table 11

Compound No.	Index of Control 500 ppm
6	3
21	3
27-b	3
34	3
51-b	3
53-b	3
104	3

Test Example 10

Test on preventive effect against oat crown rust

Oats (cultivars: Zenshin) were cultivated in a polyethylene pot having a diameter of 7.5 cm. When oats reached the two-leaf stage, 10 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration was sprayed over oats using a spray gun. After keeping the pots in a constant temperature chamber of 22 to 24° C over one day and one night, conidia of fungi of crown rust (Puccinia coronata) were inoculated. Ten days after the inoculation, an area of lesion on the second leaf was investigated, and an index of control was determined by the standards for evaluation described above. The results shown in Table 12 were obtained.

Table 12

Compound No.	Index of Control 500 ppm
4	3
14-b	3
44	4
52	4
59-b	3
104	4
172-b	4
180	5
190	3

Test Example 11

Test on preventive effect against turnip clubroot

Soil contaminated with fungi of clubroot (Plasmodiophora brassicae) was filled in a 1/14,000 a (1/140 m^2) pot, and 20 ml of a solution obtained from each of test compounds adjusted to 4 kg/10 a and 1 kg/10 a calculated as the active ingredient was drenched on the surface of the soil using a pipette. One day after treatment, the soil was mixed over the whole layers, and turnip (cultivars: Kanamachi Kokabu) was seeded. The turnip was grown in a greenhouse. Thirty days after the seeding, a degree of clubroot formation was investigated, and an index of control was determined by the standards for evaluation described below. The results shown in Table 13 were obtained.

Standards for Evaluation

[Index of Control]	[Degree of Occurrence of Clubroot]		
5:	formation of clubroot	:	none
4:	"	:	slight
3:	"	:	medium
2:	"	:	many
1:	"	:	abundant

Table 13

Compound No.	Index of Control 4 kg/10 a	1 kg/10 a
1	5	-
4	4	-
5	5	5
6	5	5
7	5	5
8	5	5
9-b	5	4
10-b	-	5
12	-	5
14-b	5	5
15-b	-	5
16-b	5	5
17	-	5
17-b	-	5
18	5	5
18-b	-	5
19	5	5
20-b	5	4
21	5	5
22	5	5
23	5	5
23-a	-	4
23-b	-	4
24-b	-	5

Table 13 (cont'd)

Compound No.	Index of Control 4 kg/10 a	1 kg/10 a
26	5	4
26-b	-	5
27-b	5	5
29-b	-	5
30-b	-	5
32-b	-	5
33	5	5
33-b	-	5
34	5	5
36	5	5
37	5	5
39-b	-	5
40-b	-	5
42-a	-	5
42-b	-	5
46-a	5	-
49	-	4
50	5	5
51	5	5
51-b	5	5
52	-	5
53-b	5	4
55-b	-	5
56-b	-	5

Table 13 (cont'd)

Compound No.	Index of Control 4 kg/10 a	1 kg/10 a
58-b	-	5
59-b	5	5
65-b	-	5
67-b	-	5
68-b	-	5
73-b	4	-
88	-	4
105	4	-
106	5	-
180	5	5
201-b	-	5
202-b	-	5
206-b	-	5
207-b	-	5

Test Example 12

Antimicrobial test (phytopathogenic fungi)

Mycelial disc (agar punching) of preincubated Pythium aphanidermatum was transplanted on potato-dextrose agar medium (PDA medium) containing 100 ppm of streptomycin and 100 ppm of each of test compounds. After incubation at 22°C for 48 hours, a diameter of mycelium was measured. Inhibition of hyphal growth (%) was determined by the following equation. The results shown in Table 14 were obtained.

$$\textit{inhibition}\ \text{of hyphal growth}\ (\%) = 100 - \frac{\text{Diameter of } \textit{mycelium} \text{ in treated plot}}{\text{Diameter of } \textit{mycelium} \text{ in non-treated plot}} \times 100$$

Table 14

Compound No.	Inhibition of Hyphal Growth (%)
3-b	100
5	95
7	100
9-b	100
10-b	100
14-b	100
15-b	100
16-b	100
17	100
17-b	100
23	100
27-b	100
28-b	100
29-b	100
30-b	100
31	100
33	100

Table 14 (cont'd)

Compound No.	Inhibition of Hyphal Growth (%)
34	100
36	100
37	100
45	100
49	100
53-b	100
101	100
103	100
104	100
105	100
106	100
180	100

Test Example 13

Miticidal test on adults of two-spotted spider mites

Kidney bean (cultivars: Edogawa Saito) was cultivated in a polyethylene pot having a diameter of 7.5 cm. When kidney bean reached the primary leaf stage, one primary leaf was left, and other leaves were cut out. After infesting about 30 adults of two-spotted spider mite (Tetranychus urticae: resistant to Dicofol and organic phosphorus insecticides), the seedlings were immersed in 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration for about 10 seconds. After drying, the seedlings were allowed to stand in a constant temperature chamber of 26°C with lighting. Two days after releasing the mites, numbers of dead mites were investigated, and a mortality (%) was determined by the following equation. The results shown in Table 15 were obtained.

$$Mortality\,(\%) = \frac{\text{Number of dead } mites}{\text{Number of released } mites} \times 100$$

Table 15

Compound No.	Mortality (%) 800 ppm	200 ppm
9-b	100	100
10-b	100	100
14-b	100	100
15-b	100	100
23	100	100
23-a	100	-
23-b	91	-
26-b	100	-
29-b	100	100
36	100	100
40-b	100	100
41	100	-
52	100	100

Table 15

Compound No.	Mortality (%) 800 ppm	200 ppm
57-b	100	-
58-b	100	-
72	100	-
88	100	-
101	100	100
112-b	100	-
113-b	100	100
119-b	100	100
133-b	100	90
151-b	100	-
167-b	100	87
169-b	100	100
172-b	100	-
205-b	100	-

Test Example 14

Ovicidal test on two-spotted spider mites

Kidney bean with only one primary leaf was transplanted on a polyethylene pot. After infesting adults of two-spotted spider mite (Tetranychus urticae) and ovipositing for 24 hours, the adults were removed. Then, the kidney bean described above was immersed in 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration for about 10 seconds. After drying, the kidney bean was kept in a constant temperature chamber of 26° C with lighting. Five to seven days after the treatment, a state of hatching was investigated, and an ovicidal rate (%) was determined by the following equation. The results shown in Table 16 were obtained. Death immediately after hatching was regarded to be ovicidal.

$$\text{Ovicidal Rate (\%)} = \frac{\text{Number of killed } eggs}{\text{Number of oviposited } eggs} \times 100$$

Table 16

Compound No.	Ovicidal Rate (%) 800 ppm
10-b	100
15-b	100
26-b	100
29-b	100
30-b	70
40-b	100
52	98
57-b	90
88	100
101	100
113-b	100
119-b	100
133-b	100
167-b	100
169-b	100

Test Example 15

Insecticidal test on small brown planthoppers

Young seedlings of rice plant were immersed in 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration for about 10 seconds. After drying, the root was wrapped with wet absorbent cotton and put in a test tube. Then, 10 larvae of second to third instar of small brown planthoppers (Laodelphax striatellus) were released in the test tube, and the opening of the test tube was covered with gauze. The test tube was kept in a constant temperature chamber of 26° C with lighting. Five days after the release of the larvae, numbers of dead insects were investigated, and a mortality rate (%) was determined by the following equation. The results shown in Table 17 were obtained.

$$Mortality\,(\%) = \frac{\text{Number of dead } insects}{\text{Number of released } insects} \times 100$$

Table 17

Compound No.	Mortality (%) 800 ppm	200 ppm
14-b	100	100
15-b	100	80
40-b	100	-
113-b	100	-
119-b	100	-
133-b	100	-
151-b	100	-

Test Example 16

Insecticidal test on green peach aphids

A piece of cabbage leaf was immersed in 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration for about 10 seconds, followed by drying. Wet filter paper was put on a petri dish having a diameter of 9 cm, and the air-dried leaf piece was put thereon. Apterous viviparous females of green peach aphids (Myzus persicae) were released on the leaf. The petri dish was covered and kept in a constant temperature chamber of 26° C with lighting. Two days after release of the insects, numbers of dead insects were investigated, and a mortality (%) was determined in the same manner as Test Example 15 described above. The results shown in Table 18 were obtained.

Table 18

Compound No.	Mortality (%) 800 ppm
1	70
32-b	70
52	90

Test Example 17

Insecticidal test on common cutworms

A piece of cabbage leaf was immersed in 20 ml of a solution obtained from each of test compounds adjusted to a predetermined concentration for about 10 seconds followed by drying. Wet filter paper was put on a petri dish having a diameter of 9 cm, and the air-dried leaf piece was put thereon. Second to third instar larvae of common cutworms (Spodoptera litura) were released on the leaf. The petri dish was covered and kept in a constant temperature chamber of 26° C with lighting. Five days after release of the larvae, numbers of dead insects were investigated, and a mortality (%) was determined in the same manner as Test Example 15 described above. The results shown in Table 19 were obtained.

Table 19

Compound No.	Mortality (%) 800 ppm
26-b	100
40-b	100
67-b	100
68-b	100
72	100
74	100

Test Example 18

Antimicrobial test (fungi)

Trichophyton metagrophytes and Trichophyton rubrum were inoculated on Sabouraud agar medium containing 10 ppm of kanamycin and each of test compounds. After incubation at 28 to 30° C for 5 days, growth of text fungi was examined. As the results, Compound Nos. 25, 34, 55-b, 119-b, and 168-b were effective against Trichophyton metagrophytes, and Compound No. 23 was effective against Trichophyton rubrum. Compound Nos. 26, 120-b, 134, and 169-b were effective against both fungi.

Test Example 19

Antimicrobial test (bacteria)

Staphylococcus aureus was inoculated on bouillon agar medium containing 10 ppm of each of test compounds. After incubation at 37° C for 16 hours, growth of test bacteria was examined. As the results, Compound Nos. 17, 20-b, 21, 22, 23, 25, 26, 26-b, 28-b, 33, 34, 37, 41, 42-a, 43, 57-b, 67-b, 103, 104, 105, 106, 134, 168-b, 201-b, 202-b, 203-b, and 205-b were effective.

Formulation examples of the present invention are described below, but the compounds, dose in formulations, type of formulations, etc. in the present invention are not deemed to be limited to those described below.

Formulation Example 1 (Wettable powder)

(a) Compound No. 5 50 parts by weight
(b) Kaolin 40 parts by weight
(c) Sodium lignin sulfonate 7 parts by weight
(d) Dialkylsulfosuccinate 3 parts by weight
The above components are uniformly mixed.

Formulation Example 2 (Wettable powder)

(a) Compound No. 17-b 20 parts by weight
(b) Kaolin 72 parts by weight
(c) Sodium lignin sulfonate 4 parts by weight
(d) Polyoxyethylene alkylaryl ether 4 parts by weight
The above components are uniformly mixed.

Formulation Example 3 (Wettable powder)

(a) Compound No. 18-b 6 parts by weight
(b) Diatomaceous earth 88 parts by weight
(c) Dialkylsulfosuccinate 2 parts by weight
(d) Polyoxyethylene alkylaryl sulfate 4 parts by weight
The above components are uniformly mixed.

Formulation Example 4 (Wettable powder)

(a) Kaolin 78 parts by weight
(b) Sodium β-naphthalene-sulfonate-formaldehyde condensate 2 parts by weight
(c) Polyoxyethylene alkylaryl sulfate 5 parts by weight
(d) Fine silica 15 parts by weight
A mixture of the above components and Compound No. 22 are mixed in a weight ratio of 4:1.

Formulation Example 5 (Wettable powder)

(a) Compound No. 16-b 10 parts by weight
(b) Diatomaceous earth 69 parts by weight
(c) Calcium carbonate powder 15 parts by weight
(d) Dialkylsulfosuccinate 1 part by weight
(e) Polyoxyethylene alkylaryl sulfate 3 parts by weight
(f) Sodium β-naphthalene-sulfonate-formaldehyde condensate 2 parts by weight
The above components are uniformly mixed.

Formulation Example 6 (Wettable powder)

(a) Compound No. 17-b 20 parts by weight
(b) Kaolin 62.4 parts by weight
(c) Fine silica 12.8 parts by weight
(d) Alkylaryl sulfonate 1.6 parts by weight
(e) Polyoxyethylene alkylaryl sulfate 2.4 parts by weight
(f) Polyoxyethylene alkylaryl ether 0.8 parts by weight
The above components are uniformly mixed.

Formulation Example 7 (Dust)

(a) Compound No. 23 5 parts by weight
(b) Talc 94.5 parts by weight
(c) Lower alcohol phosphate 0.5 parts by weight
The above components are uniformly mixed.

Formulation Example 8 (Dust)

(a) Compound No. 16-b 0.2 parts by weight
(b) Calcium carbonate powder 98.8 parts by weight
(c) Lower alcohol phosphate 1.0 parts by weight
The above components are uniformly mixed.

Formulation Example 9 (Emulsifiable concentrate)

(a) Compound No. 26 20 parts by weight
(b) Xylene 60 parts by weight
(c) Polyoxyethylene alkylaryl ether 20 parts by weight
The above components are mixed and dissolved.

Formulation Example 10 (Suspension concentrate)

(a) Compound No. 151 10 parts by weight
(b) Corn oil 77 parts by weight
(c) Polyoxyethylene caster oil 12 parts by weight
(d) Bentonite-alkylamine complex 1 part by weight
The above components are uniformly mixed and pulverized.

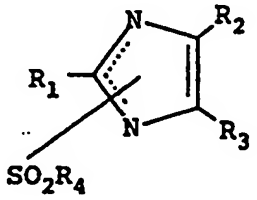
Formulation Example 11 (Granule)

(a) Compound No. 33-b 1 part by weight
(b) Bentonite 61 parts by weight
(c) Kaolin 33 parts by weight
(d) Sodium lignin sulfonate 5 parts by weight
A suitable amount of water required is added to the above components, followed by mixing and granulating.

While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

Claims

1. An imidazole compound represented by the following general formula (I):



(I)

wherein:

R_1 represents a cyano group or a -CSNHR$_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a -COR$_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an -SO$_n$R$_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an -NR$_8$R$_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a -CO(NH)$_m$R$_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an -NR$_{11}$R$_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom.

2. The compound according to Claim 1, wherein R_1 represents a cyano group.

3. The compound according to Claim 1, wherein R_1 represents a cyano group; R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, or nitro groups; an SO$_n$R$_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a phenyl group which is optionally substituted with one or more halogen atoms, or an -NR$_8$R$_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1 or 2; or a -CONHR$_{10}$ group, wherein R_{10} represents a phenyl group which is optionally substituted with one or more halogen atoms; and R_4 represents a C_{1-6} alkyl group or an -NR$_{11}$R$_{12}$ group, wherein R_{11} and R_{12} each represents a C_{1-4} alkyl group; provided that R_2 and R_3 are not simultaneously a halogen atom.

4. The compound according to Claim 1, wherein R_1 represents a cyano group; R_2 represents a hydrogen atom; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, phenyl groups, or halogenated phenyl groups; a C_{2-4} alkenyl group; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, C_{1-4} alkoxy groups, or halogenated C_{1-4} alkoxy groups; a C_{1-6} alkylthio group; or a phenylthio group which is optionally substituted with one or more halogen atoms; R_3 represents a hydrogen atom; a halogen atom; or a cyano group; and R_4 represents an -N(CH$_3$)$_2$ group.

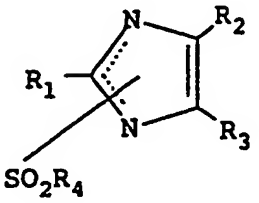
5. The compound according to Claim 1, wherein R_4 represents an N(CH$_3$)$_2$ group.

6. The compound according to Claim 1, wherein R_1 represents a cyano group, and R_4 represents an -N(CH$_3$)$_2$ group.

7. The compound according to Claim 1, wherein R_1 represents a cyano group; R_2 represents a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, phenyl groups, or halogenated phenyl groups; a C_{2-4} alkenyl group; a phenyl group which is optionally substituted with one or more halogen atoms; or a C_{1-6} alkylthio group; R_3 represents a halogen atom; and R_4 represents an N(CH$_3$)$_2$ group.

8. The compound according to Claim 1, wherein R_1 represents a cyano group; R_2 represents a C_{1-12} alkyl group or a phenyl group; R_3 represents a chlorine atom; and R_4 represents an -N(CH$_3$)$_2$ group.

9. A biocidal composition which comprises an imidazole compound, as an active ingredient, represented by the following general formula (I):



(I)

wherein:

R_1 represents a cyano group or a $CSNHR_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a $-COR_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

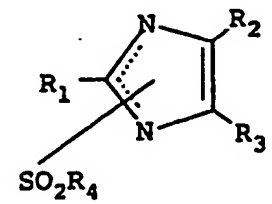
R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an $-NR_{11}R_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom;

and adjuvants.

10. A process for preparing an imidazole compound represented by the following general formula (I):



(I)

wherein:

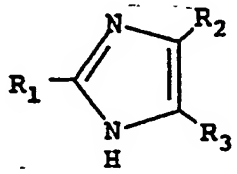
R_1 represents a cyano group or a $-CSNHR_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a $-COR_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms; a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an $-NR_{11}R_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom;

which comprises reacting a compound represented by the following general formula (II):



(II)

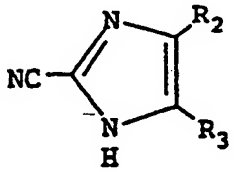
wherein R_1, R_2, and R_3 are as defined above,

with a compound represented by general formula (III):

$Y\text{-}SO_2R_4$ (III)

wherein R_4 is as defined above, and Y represents a halogen atom.

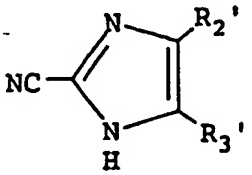
11. A compound as an intermediate represented by the following general formula (II'):



(II')

wherein R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one

or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4} alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1;
provided that compounds represented by the general formula (II"):

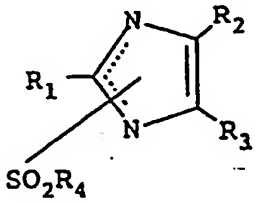


(II")

wherein R_2' and R_3' are simultaneously a hydrogen atom, a halogen atom, a cyano group, or a phenyl group which is optionally substituted with the same or different C_{1-2} alkoxy group or C_{1-2} alkylthio group at the para-position; and wherein R_2' is a hydrogen atom and R_3' is a methyl group or a phenyl group,
are excluded.

Claims for the following Contracting State : ES

1. An agricultural and/or horticultural biocidal composition for controlling organisms harmful to plants, which comprises from 0.05 to 90 parts by weight of an imidazole compound, as an active ingredient, represented by the following general formula (I):



(I)

wherein:

R_1 represents a cyano group or a $-CSNHR_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a $-COR_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a trimethylsilyl group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-12} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, acetoxy groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, phenyl groups, halogenated phenyl groups, or C_{1-4} alkylated phenyl groups; a C_{2-10} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, cyano groups, or 3,4-methylenedioxy groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a phenyl group, which is optionally substituted with one or more halogen atoms, a benzyl group, a pyridyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, or halogenated C_{1-4}

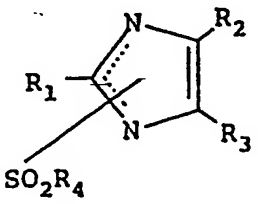
alkyl groups; or an $-NR_8R_9$ group, wherein R_8 and R_9 each represents a C_{1-4} alkyl group, and n is 0, 1, or 2; or a $-CO(NH)_mR_{10}$ group, wherein R_{10} represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{1-4} alkoxy group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and m is 0 or 1; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an $-NR_{11}R_{12}$ group, wherein R_{11} and R_{12} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_{11} and R_{12} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_{11} and R_{12} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom;

and from 10 to 99.95 parts by weight of at least one kind of agriculturally acceptable adjuvants.

2. The biocidal composition according to Claim 1, wherein an imidazole compound, as an active ingredient, is represented by the following general formula:



wherein:

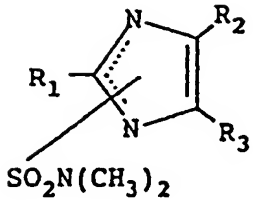
R_1 represents a cyano group, or a $-CSNHR_5$ group, wherein R_5 represents a hydrogen atom, a C_{1-4} alkyl group, or a $-COR_6$ group, wherein R_6 represents a C_{1-4} alkyl group, a halogenated C_{1-4} alkyl group, or a phenyl group;

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a nitro group; a cyano group; a C_{3-6} cycloalkyl group; a naphthyl group; a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms, hydroxyl groups, C_{1-4} alkoxy groups, phenyl groups, or halogenated phenyl groups; a C_{2-6} alkenyl group which is optionally substituted with one or more halogen atoms; a C_{1-6} alkoxy group; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups, C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, or cyano groups; a furyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; a pyridyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; an $-SO_nR_7$ group, wherein R_7 represents a C_{1-6} alkyl group, a C_{2-6} alkenyl group, or a phenyl group which is optionally substituted with one or more halogen atoms, and n is 0, 1 or 2; or a $-COR_8$ group, wherein R_8 represents a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, or a phenyl group which is optionally substituted with one or more halogen atoms; and

R_4 represents a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms; a C_{3-6} cycloalkyl group; a phenyl group; a thienyl group; or an $-NR_9R_{10}$ group, wherein R_9 and R_{10} each represents a hydrogen atom, a C_{1-4} alkyl group which is optionally substituted with one or more halogen atoms, a C_{2-4} alkenyl group, or R_9 and R_{10} are combined with each other together with a nitrogen atom adjacent thereto to form a pyrrolidinyl group, a piperidinyl group, a morpholino group, or a thiomorpholino group, provided that R_9 and R_{10} are not simultaneously a hydrogen atom;

provided that R_2 and R_3 are not simultaneously a halogen atom.

3. The biocidal composition according to Claim 1, wherein an imidazole compound, as an active ingredient, is represented by the following general formula:



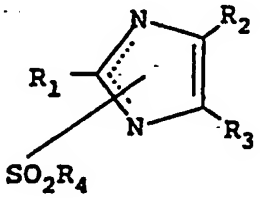
wherein:

R_1 represents a cyano group or a -$CSNH_2$ group; and

R_2 and R_3 each represents a hydrogen atom; a halogen atom; a cyano group; a naphthyl group; a C_{1-6} alkyl group which is optionally substituted with one or more halogen atoms or phenyl groups; a C_{2-6} alkenyl group which is optionally substituted with one or more halogen atoms; a phenyl group which is optionally substituted with one or more halogen atoms, C_{1-4} alkyl groups, halogenated C_{1-4} alkyl groups, C_{1-4} alkoxy groups, halogenated C_{1-4} alkoxy groups C_{1-4} alkylthio groups, halogenated C_{1-4} alkylthio groups, nitro groups, or cyano groups; a thienyl group which is optionally substituted with one or more halogen atoms or C_{1-4} alkyl groups; or an -SR_4 group, wherein R_4 represents a C_{1-6} alkyl group or a phenyl group which is optionally substituted with one or more halogen atoms;

provided that R_2 and R_3 are not simultaneously a halogen atom.

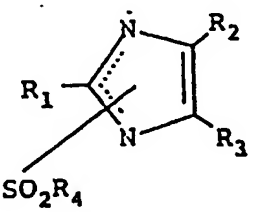
4. A method of controlling organisms harmful to plants comprising the step of treating the organisms and/or plants and/or earth with a biocidal composition comprising an imidazole compound represented by the following general formula (I):



(I)

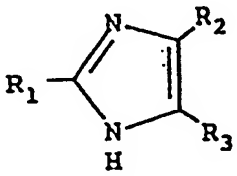
wherein R_1, R_2, R_3 and R_4 are as defined in any of Claims 1 to 3.

5. A process for preparing a compound of general formula (I):



(I)

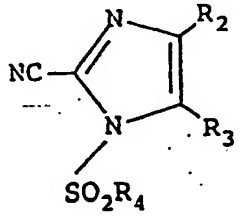
wherein R_1, R_2, R_3 and R_4 are as defined above comprising the step of reacting Y-SO_2R_4 wherein Y is a halogen atom and R_4 is as defined above with a compound of general formula (II):



(II)

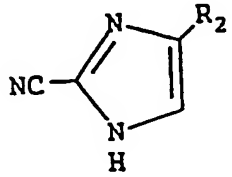
wherein R_1, R_2 and R_3 are as defined above.

6. A process for preparing a compound of general formula (I-2):



(I-2)

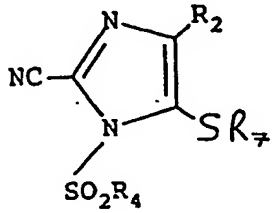
wherein R_2, R_3 and R_4 are as defined above comprising the steps of reacting Y-SO_2R_4 wherein Y is a halogen atom and R_4 is as defined above with a compound of general formula (II-1)



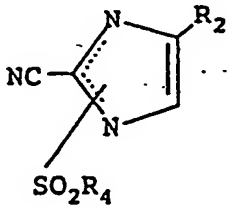
(II-1)

wherein R_2 is as defined above, and
reacting the product thereof with a compound of formula R_3I wherein R_3 is as defined above.

7. A process for preparing a compound of general formula:



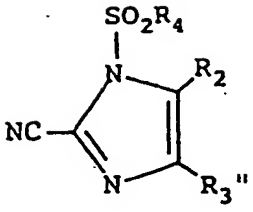
wherein R_2, R_4 and R_7 are as defined above comprising the step of reacting a compound of formula R_7SSR_7 wherein R_7 is as defined above with a compound of general formula (I-1):



(I-1)

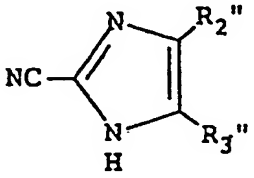
wherein R_2 and R_4 are as defined above.

8. A process for preparing a compound of general formula (I-4):



(I-4)

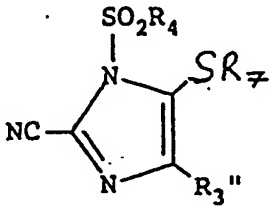
wherein R_2 and R_4 are as defined above and R_3'' is a hydrogen atom, a chlorine atom or a bromine atom, comprising the steps of reacting a compound of formula $Y\text{-}SO_2R_4$ wherein Y is a halogen atom and R_4 is as defined above with a compound of general formula (II-2).



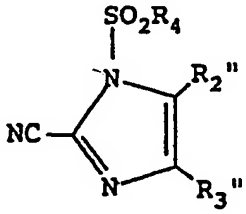
(II-2)

wherein R_3'' is as defined above and R_2'' represents the same substituent as R_3''.
and reacting the product thereof with R_2Y' wherein R_2 is as defined above and Y' is a chlorine atom, bromine atom or an iodine atom.

9. A process for preparing a compound of formula

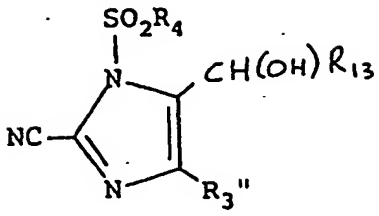


wherein R_4, R_3'' and R_7 are as defined above comprising the step of reacting R_7SSR_7 wherein R_7 is as defined above with a compound of formula

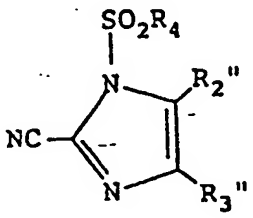


wherein R_4 and R_3'' are as defined above and R_2'' represents the same substituent as R_3''.

10. A process for preparing a compound of formula

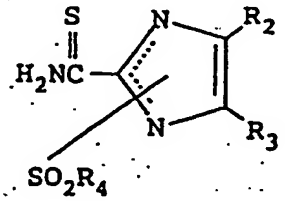


wherein R_3'' and R_4 are as defined above and R_{13} is an alkyl group or an optionally substituted phenyl group comprising the step of reacting $R_{13}CHO$ wherein R_{13} is as defined above with a compound of formula



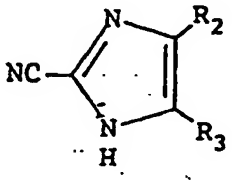
wherein R_4 and R_3'' are as defined above and R_2'' represents the same substituent as R_3''.

11. A process for preparing a compound of general formula (I-6):



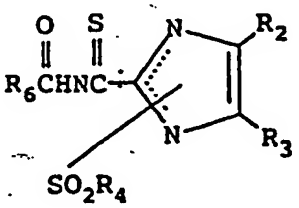
(I-6)

wherein R_2, R_3 and R_4 are as defined above comprising the step of reacting YSO_2R_4 wherein Y is a halogen atom and R_4 is as defined above with a compound of formula:



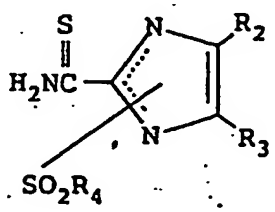
wherein R_2 and R_3 are as defined above,
and reacting the product with H_2S.

12. A process for preparing a compound of general formula (I-7):



(I-7)

wherein R_2, R_3, R_4 and R_6 are as defined above comprising the step of reacting R_6COCl wherein R_6 is as defined above with a compound of general formula (I-6):



(I-6)

wherein R_2, R_3 and R_4 are as defined above.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 88 10 3885

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	CHEMICAL ABSTRACTS, vol. 92, no. 21, 26th May 1980, page 656, column 2, abstract no. 181177f, Columbus, Ohio, US; & JP - A - 79 90175 (NISSAN CHEMICAL INDUSTRIES LTD.) 17-07-1979	1,9,10	C 07 D 233/90 C 07 D 233/92 C 07 D 405/04 C 07 D 409/04 C 07 D 401/04 C 07 D 401/12 C 07 D 409/12 C 07 D 405/14 C 07 F 7/08 A 01 N 43/56 A 61 K 31/415 A 61 K 31/44 // C 07 D 233/64 C 07 D 233/58
A,D	EP-A-0 031 086 (BAYER AG) * claims 1,4-7; examples 7,14 *	1,9,11	
A	US-A-4 574 010 (A. LEONE-BAY et al.) * whole document *	1,9,10	
A	US-A-4 220 466 (N.R. PATEL) * column 1, lines 42-68; example 42 *	1,9	
A	DE-A-1 770 850 (MERCK & CO. INC.) * claims 1,5; page 4, lines 18-21 *	1,9	
A	DE-A-2 634 053 (BAYER AG) * claims 1,7-10, examples 119,128 *	1,9,11	
A	CHEMICAL ABSTRACTS, vol. 105, no. 3, 21st July 1986, page 288, column 2, abstract no. 20528t, Columbus, Ohio, US; & US - A - 759 856 (UNITED STATES DEPARTMENT OF ENERGY) 14-03-1986	1,9	TECHNICAL FIELDS SEARCHED (Int. Cl.4) C 07 D 233/00 C 07 D 405/00 C 07 D 409/00 C 07 D 401/00 A 01 N 43/00 A 61 K 31/00
P,A D	CHEMICAL ABSTRACTS, vol. 107, no. 19, 9th November 1987, page 726, column 2, abstract no. 176038k, Columbus, Ohio, US; & JP - A - 62 142 164 (ISHIHARA SANGYO KAISHA, LTD.) 25-06-1987	1,9,10	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	23-06-1988	VAN AMSTERDAM L.J.P.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

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